

NASA TT F-11,623

STUDY OF THERMOCOUPLES FOR MEASUREMENT OF HIGH TEMPERATURES

Michel Villamayor

Translation of *Etude de Thermocouples pour le Repérage des Hautes Températures*,
a thesis presented to the Science Faculty of the University
of Lyon for the degree of Doctor of Applied Sciences

and published by the Documentation Service of

the Commissariat à l'Energie Atomique,
Centre d'Etudes Nucléaires de Saclay,

as CEA Report R-3182

JAN. 1967, 111 pp

FACILITY FORM 602

N68-22383

(ACCESSION NUMBER)

84

(PAGES)

(THRU)

(CODE)

14

(CATEGORY)

(NASA CR OR TMX OR AD NUMBER)



GPO PRICE \$

CFSTI PRICE(S) \$

Hard copy (HC) 3.00

Microfiche (MF) .65

ff 653 July 65

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON, D.C. 20546

APRIL 1968

FOREWORD

This work was done in the laboratory for the physics of matter at the Institut National des Sciences Appliquées at Lyon and in the reactor service of the Centre d'Etudes Nucléaires at Grenoble. I hereby ask Dean H. Lefebvre, Director of the INSA, Lyon; Professor L. Neel, Director of the CEN, Grenoble; and Mr. Salesse, head of the department of metallurgy of the Commissariat à l'Energie Atomique to accept my respectful acknowledgement of their material and financial contributions, without which nothing would have been done.

I very warmly thank Professor Ch. Eyraud, head of the department of mechanics of the INSA at Lyon, for doing me the honor of accepting the chairmanship of the examination committee.

Let me express here my deep and sincere gratitude to Professor F. Davoine, now Director of the Ecole Nationale Supérieure de la Métallurgie et de l'Industrie des Mines, Nancy, who, after having directed my undergraduate engineering studies, has guided and watched over my first steps in research. His frequent advice, suggestions, and friendly criticisms have enabled me to carry out this work in an atmosphere to which he was always able to impart cordiality. Mr. P. Pinard, lecturer, has made it possible for me in the last phases of this study to profit fully from his experience, and I am happy to be able to express to him here my very warm gratitude.

An important part of this work has been made possible by the collaboration of Mr. F. Rossillon, head of the reactor service of the CEN, Grenoble. Let me assure him of my most sincere thanks.

It was Mr. P. Thome, head of the advanced techniques section of the CEN, Saclay, who was responsible for directing the various stages of this study. I hereby express to him my feeling of deep gratitude.

I also thank all my colleagues and the personnel of the laboratory for their constant collaboration and the many kindnesses that they have always shown me.

TABLE OF CONTENTS

	Page
Introduction	1
Part One	
A. Theoretical Review of Thermoelectricity	3
I. Thermoelectric Effects	3
II. Interpretation of the Thermoelectric Effects; Thomson Relations	4
III. Laws of Thermoelectric Effects	6
IV. Principles of the Use of Thermocouples	6
B. Thermocouples	9
I. Selection of Thermocouples	9
II. Thermocouples of Refractory Metals and Alloys	9
III. Thermocouples of Tungsten-Rhenium Alloys	10
Part Two. Study of Tungsten-Rhenium Alloy Thermocouples Outside the Reactor	
A. Preliminary Studies	14
I. Experimental Apparatus	14
II. Statistical Calibrations; Dispersion; Mean Calibration Curves	20
III. Stability in the Course of Time	23
IV. Stability in Gases	25
B. Supplementary Studies	27
I. Experimental Apparatus	27
II. Influence of Thermal Shocks	27
III. Measurement of the Response Time	33
C. Study of Cold Welding Compensation	36
I. Experimental Apparatus	36
II. Study of the Alloys	36
III. Comparison with X-Y Wires	37
Part Three. Study of Tungsten-Rhenium Alloy Thermocouples In the Reactor	
A. Experimental Arrangement	40
I. The Reactor SILOE and the Furnace HEBE	40
II. The Calibration Furnace TETARD	43
III. The Sheaths Containing the Thermocouples	45
IV. The Auxiliary Devices	47
B. Experimentation	
I. General	48
II. The Precalibrations	49
III. The Irradiations	50

	Page
IV. Calibrations After Irradiation	52
C. Results Obtained	53
I. Preliminary Remarks	53
II. Recordings During the Calibrations	53
III. Curves of Deviation	56
IV. Conclusion	56
D. Influential Factors	57
I. Transmutations	57
II. Perturbation of the Lattice	60
III. Other Factors	61
Appendix. Behavior of Thermocouples of Refractory Alloys Under Irradiation in the Presence of Uranium Dioxide	62
Conclusion	64
Tables and Mean Calibration Curves of Tungsten-Rhenium Alloy Thermocouples	67
References	75

STUDY OF THERMOCOUPLES FOR MEASUREMENT OF HIGH TEMPERATURES

Michel Villamayor

ABSTRACT. Previous works have shown that tungsten-rhenium alloy thermocouples were a good instrument for measuring high temperatures. Starting with that fact the author has studied the W/W 26% Re and W 5% Re/W26% Re thermocouples of French manufacture intended for measuring temperatures up to 2300°C. In experiments outside the reactor he has determined the general characteristics of these thermocouples -- average calibration curves, thermal shock influence, response times, and alloys which permit cold source compensation. The evolution of these thermocouples under thermal neutron flux has been determined by experiments in the reactor. The observations have led the author to propose a new type of thermocouple made of molybdenum-columbium alloys.

INTRODUCTION

/5*

Measurement of high temperatures is generally done in one of two ways: either by means of an optical pyrometer, or by means of probes.

The first method makes it possible to reach very high temperatures, but while its use is valuable, there are a number of cases where it cannot be used.

As to the second method, until the last few years it had been limited to a restricted range of temperatures, since the thermocouples were made of precious metals.

It was not until the recent improvement in processes of manufacture of refractory metals and alloys that it was possible to produce rugged thermocouples of small bulk capable of precise measurement of very high temperatures, and in particular the operating temperatures of nuclear reactors.

The measurement of the electromotive forces provided by numerous combinations of refractory metals and alloys has been carried out in foreign laboratories, and the results obtained show that it is tungsten-rhenium alloys that offer the best thermoelectric characteristics.

In this study we have investigated the behavior of thermocouples made of pure tungsten and tungsten with 26% rhenium (W/W 26% Re) and of tungsten with 5% rhenium and tungsten with 26% rhenium (W 5% Re/W 26% Re), of French manufacture (Société J. Bocuze & Cie), designed for measuring the tempera-

*Numbers in the margin indicate pagination in the foreign text.

ture in nuclear reactors.

The general characteristics of these thermocouples were determined in experimentation outside the reactor.

By calibrating a large number of them we were able to establish the /6 curves and mean calibration tables relative to each type.

We then attempted to delimit the errors in measurement resulting from the conditions of their use, investigating their response time, their variation as a function of time, and the influence of thermal shocks.

In order to take account, within a wide range of temperature, of the evolution of these thermocouples under irradiation, we undertook a series of manipulations, the description and the results of which are the subject of the study in the reactor.

The observed phenomena, which can be interpreted quite well by attributing an important rôle to the effects produced by nuclear transmutations, have led us to propose a new type of thermocouples made of molybdenum-niobium alloys.

A. Theoretical Review of Thermoelectricity

B. Thermocouples

A. THEORETICAL REVIEW OF THERMOELECTRICITY

I. THERMOELECTRIC EFFECTS

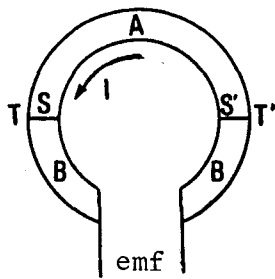
1) *The Seebeck Effect* (1821)

Figure 1

Let us consider the circuit represented by Figure 1 and consisting of two homogeneous conductors A and B in contact along two surfaces S and S' kept at the temperatures T and T' respectively.

In an open circuit we observe the appearance of an electromotive force (emf) dependent on the nature of the conductors and on the temperatures T and T'. In a closed circuit this emf induces the passage of a current which is reversed when T and T' are transposed.

We shall denote the emf by $E_{T,T'}^T$, (A,B), with $T > T'$ and the current flowing in the direction indicated on the figure.

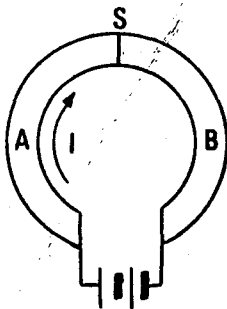
2) *The Peltier Effect* (1834)

Figure 2

Assume an electric circuit consisting of two homogeneous conductors A and B, in contact along a surface S and kept at a temperature T, in which a current of the intensity I is made to circulate (Figure 2). At the surface S we observe a release or an absorption of heat such that

$$\frac{dQ}{dt} = P_{ab} \cdot I .$$

P_{ab} is called Peltier's coefficient or Peltier's /10 voltage, and depends on the nature of the conductors and on T.

3) The Thomson Effect

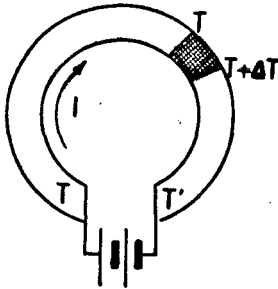


Figure 3

Let us imagine a homogeneous conductor carrying a current of the intensity I , the extremities of which are kept at temperatures T and T' (Figure 3). In a region where the temperature passes from T to $T + \Delta T$, we observe an emission or an absorption of heat such that

$$\frac{dQ}{dt} = h \cdot I \cdot \Delta T .$$

In the entire conductor we shall then have

$$\frac{dQ}{dt} = I \cdot \int_{T'}^T h \cdot dT ,$$

where h is Thomson's coefficient.

In order to simplify our later calculations we shall write H_T^T , for the integral $\int_{T'}^T h \cdot dT$ which is homogeneous at a voltage and which we shall call the "Thomson voltage."

II. INTERPRETATION OF THE THERMOELECTRIC EFFECTS; THOMSON RELATIONS

In a conductor in which there exist simultaneously an electric current (intensity I), a temperature gradient ($\text{grad } T$), and a structure gradient ($\text{grad } f$, function of the concentration of free electrons and of the temperature gradient), the equations of electric flow and thermal flow are written respectively:

$$(1) \quad I = A \cdot \text{grad } f + B \cdot \frac{\text{grad } T}{T}$$

$$(2) \quad K = B \cdot \text{grad } f + C \cdot \frac{\text{grad } T}{T}$$

Equation (1) can be put in the form:

$$\text{grad } f = e(\rho \cdot I + S \cdot \text{grad } T) ,$$

with $-e$: the charge of the electron,

/11

ρ : the resistivity of the conductor, and

S : the absolute thermoelectric power of the conductor.

It is interesting to note that Mott [7] has calculated the expression relating the absolute thermoelectric power S of a pure metal to its energy characteristics.

$$S = \frac{\pi^2 \cdot k^2 \cdot T}{3e} \left(\frac{d \log \rho(E)}{dE} \right)_{E = E_F}$$

k : Boltzmann's constant; E_F : the Fermi energy.

Domenicalli [8] has since extended the formula to the case of alloys.

It has been demonstrated that the quantity of thermoelectric heat emitted per unit volume per unit time is equal to:

$$(4) \quad \frac{dQ}{dt} = T \cdot I \cdot \text{grad } S .$$

1) The Seebeck Effect

Equation (3) applied to the circuit of Figure 1 supplies the value of the thermoelectric power of the thermocouple thus constituted.

$$(5) \quad \frac{dE}{dT} = S_a - S_b .$$

2) The Peltier Effect

In the case of the circuit diagrammed in Figure 2, identification of the expressions of thermoelectric heat permits calculating the Peltier voltage:

$$(6) \quad P_{ab} = T(S_b - S_a)$$

Combining equations (5) and (6) leads to:

$$(7) \quad \frac{dE}{dT} = - \frac{P_{ab}}{T} ,$$

an equation which relates the thermoelectric power to Peltier's coefficient, and which is called "the second Thomson relation."

3) The Thomson Effect

/12

Reasoning in the same manner as before, we obtain the Thomson coefficient applied to the circuit of Figure 3:

$$h = T \frac{dS}{dT} .$$

If we extend this equation to the circuit constituting a thermocouple and take into account equation (5), we get:

$$(8) \quad h_a - h_b = T \frac{d^2 E}{dT^2} .$$

4) Expression of the Electromotive Force of a Thermocouple

As a function of the Peltier and Thomson voltages, the combination of the foregoing equations furnishes the expression of the emf delivered by a thermocouple, also called the "first Thomson relation":

$$E_{T, (A,B)}^T = P_{ab}(T) - P_{ab}(T') + H_{T, (B)}^T - H_{T, (A)}^T .$$

It must be noted that (8) imposes upon E a non-linear variation as a function of the temperature. In fact it has been found experimentally that the curve $E = f(T)$ is parabolic. It is for that reason that it is customary to represent the emf of a thermocouple by an equation of the form:

$$E = a(T - T') + b(T^2 - T'^2) + c(T^3 - T'^3) + \dots$$

It will be remembered that in $E_{T'}^T(A,B)$, by stipulation, $T > T'$, and also that A denotes the conductor which has the higher absolute thermoelectric power.

III. LAWS OF THERMOELECTRIC EFFECTS

1) Law of Metallic Chains

The sum of the Peltier effects for a metallic chain of uniform temperature is zero:

$$P_{ab} + P_{bc} + \dots + P_{na} = 0 ,$$

or

$$P_{ab} = P_{an^{\circ}} + P_{n^{\circ}b} = P_{an^{\circ}} - P_{bn^{\circ}} .$$

For the Seebeck effect this law is expressed by:

/13

$$E_{T'}^T(A,B) = E_{T'}^T(A,N_0) - E_{T'}^T(B,N_0) .$$

If the value of the Seebeck effect of two metals in respect of a reference metal is known, this relation shows that it is possible to deduce the emf delivered by the couple constituted by those two metals.

2) Law of Intermediate Temperatures

In a circuit formed of one and the same metal it is possible to analyze the Thomson voltages in such a way that:

$$H_{T^{\circ}}^T = H_{T'}^T + H_{T''}^{T'} + \dots + H_{T^{\circ}}^{T^n} .$$

The result of this for the Seebeck effect is that:

$$E_{T^{\circ}}^T(A,B) = E_{T'}^T(A,B) + E_{T^{\circ}}^{T'}(A,B) = E_{T'}^T(A,B) - E_{T'}^{T^{\circ}}(A,B) .$$

Note: In order to interpret the Seebeck effect certain authors introduce the Volta effect and the temperature effect connected with the existence of electromotive tensions inside the conductors.

Let us merely remark that these effects, which implicitly contain the Peltier and Thomson effects, obey the foregoing laws, and that their sum gives the Seebeck effect.

IV. PRINCIPLES OF THE USE OF THERMOCOUPLES

If in the circuit of Figure 1 the surface S' is kept at a reference temperature T° (0°C , for example), the measurement of the emf will make it possible to deduce the temperature T , once the function $E_{T'}^{T^{\circ}}(A,B)$ has been determined by calibration.

The law of successive temperatures authorizes us to open the circuit at any point, in this case at S' , to measure the emf, on the condition that

the two facing surfaces thus obtained are at the same temperature. In this way we shall have produced a thermocouple in its simplest form of use.

1) Measuring Device

/14

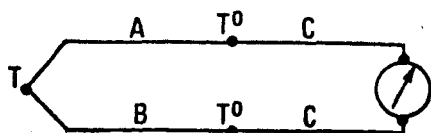


Figure 4

As the measuring instrument cannot be brought to the reference temperature (0°C), it is connected to the thermocouple by means of identical conductors, as shown in Figure 4.

The insertion of these connecting wires into the circuit introduces no modification of the emf of the thermocouple, if we assume that these wires and the materials constituting the measuring apparatus are of the same nature or simply at the same temperature.

2) Cold Welding Correction

At the terminals of a thermocouple whose cold welding is at a temperature T' different from the reference temperature T^0 with which it was calibrated, we measure $E_{T'}^T$, such that

$$E_{T'}^T = E_{T^0}^T - E_{T^0}^{T'}.$$

If to the value measured we add the emf corresponding to the temperature T' , the quantity thus obtained enables us to measure the temperature of the cold welding on the curve $E_{T^0}^T = f(T)$.

3) Cold Welding Compensation

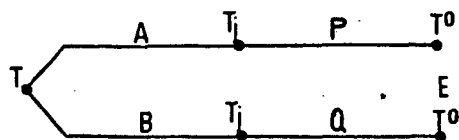


Figure 5

It is frequent that we have occasion to elongate the thermocouple by means of materials (P and Q) very different from those of which it is made, after a junction whose temperature T_j is almost always higher than the reference temperature T^0 .

Such a circuit, diagrammed in Figure 5, delivers an emf E , which we analyze into Peltier and Thomson voltages thus:

$$E = P_{qp}(T^0) + H_{T_j}^{T^0}(P) + P_{pa}(T_j) + H_T^{T_j}(A) + P_{ab}(T) + H_{T_j}^T(B) + T_{bq}(T_j) + H_{T^0}^{T_j}(Q).$$

Applying the intermediate law of metals, we get:

/15

$$P_{pa}(T_j) = P_{pq}(T_j) + P_{qa}(T_j) \quad \text{and} \quad P_{bq}(T_j) = P_{ba}(T_j) + P_{aq}(T_j),$$

and substituting in the expression for E :

$$E = [P_{ab}(T) - P_{ab}(T_j) + H_{T_j}^T(B) - H_T^{T_j}(A)] + [P_{pq}(T_j) - P_{pq}(T^0) + H_{T^0}^{T_j}(Q) - H_{T^0}^{T_j}(P)].$$

The first term enclosed in brackets represents $E_{T_j}^T(A,B)$, the second $E_{T^0}^{T_j}(P,Q)$, so that:

$$E = E_{T^0}^T(A,B) - [E_{T^0}^{T_j}(A,B) - E_{T^0}^{T_j}(P,Q)] = E_{T^0}^T(A,B) - \Delta E ,$$

ΔE being the correction to be applied in the case where the conductors P and Q are any conductors. On the other hand, if the choice of materials is such that $\Delta E = 0$, we shall then have effected the cold welding compensation.

Thus in the hypothetical case where the thermocouple made of materials P and Q delivers the same emf as the thermocouple (A,B) between T_j and T^0 , we shall in fact measure $E = E_{T^0}^T(A,B)$ at the terminals of the circuit of Figure 5.

It can also be shown that for $\Delta E = 0$, $E_{T^0}^{T_j}(A,P) = E_{T^0}^{T_j}(B,Q)$.

I. SELECTION OF THERMOCOUPLES

Of the numerous possible combinations of metals and alloys, only those which make possible the precise and efficient measurement of temperatures can be used for the manufacture of thermocouples.

Such combinations should

- deliver, within a wide range of temperature, as high and as linear an emf as possible,
- exhibit a low electrical resistivity and an excellent thermal conductivity,
- possess good mechanical strength for minimum bulk,
- be made of materials that are easily worked, so as to ensure reproducibility of production batches.

The lifetime, stability, and fidelity of thermocouples are also frequently related to the conditions under which they are used; for example, their reactivity to certain compounds makes it necessary to take precautions or even to forbid their use in the presence of such compounds; or again, thermal cycles which induce metallurgical transformations (recrystallization, tempering, etc.) within the wires contribute to a variation in the resistance and consequently in the emf.

II. THERMOCOUPLES OF REFRACTORY METALS AND ALLOYS

Measurement of temperatures up to 2300°C by means of thermocouples necessitates the use of elements of very high melting point, i.e. refractory metals (essentially tungsten, rhenium, molybdenum, niobium, and tantalum) and their alloys. Systematic studies have been undertaken [9-17] for the purpose of finding the combinations of these materials best capable of satisfying the criteria just enumerated. /19

It must be noted, however, that thermocouples with a graphite base (graphite/boron-graphite) have been developed for the same fields of application [18]. But while the emf's they deliver are very high, their use is limited by their excessive fragility and bulk, and by reason of the too great dispersion, of thermoelectric origin, that they exhibit.

1) Electromotive Force and Field of Application

Figure 6 shows the variation, as a function of the temperature, of the emf delivered by various types of thermocouple with refractory metal bases and by the Pt/Pt 10% Rh couple, which is still the most widely used for low temperatures. Comparison of the curves justifies the choice of tungsten-rhenium alloys, and in fact it will be observed that up to 2300°C the highest and most linear emf's are obtained with the combinations W/W 26% Re and W 5% Re/W 26% Re.

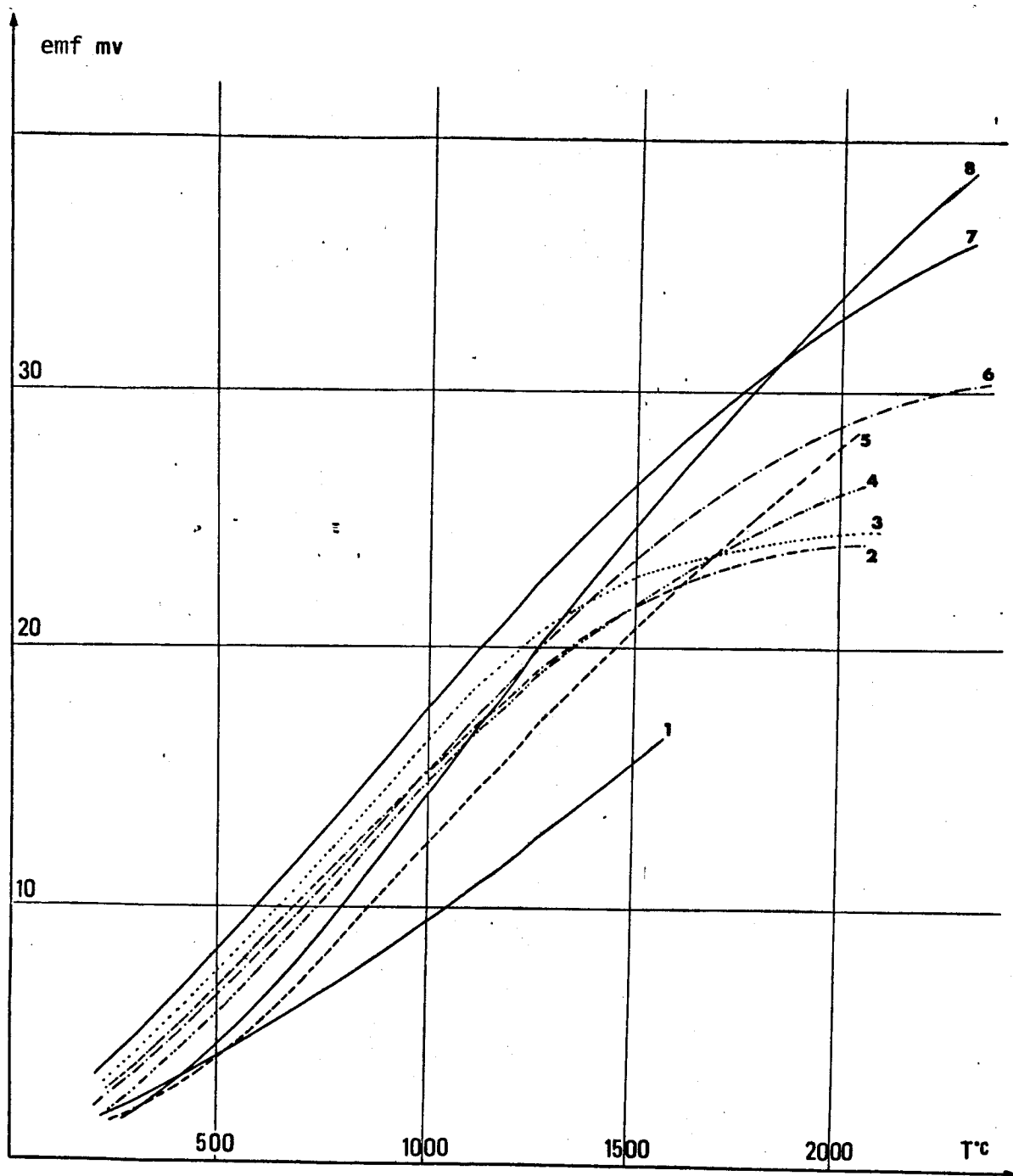


Figure 6. Curves $E_{mv} = f(T^{\circ}C)$.

/18

- 1) Pt/Pt 10% Rh
- 2) Mo/Nb
- 3) Mo/Re

- 4) Mo/Mo 50% Re
- 5) W/Nb
- 6) W/Re

- 7) W 5% Re/W 26% Re
- 8) W/W 26% Re

2) Conditions of Use

In the absence of neutron fluxes, whose influence will be the subject of a later chapter, it is primarily the chemical and mechanical properties of refractory metals and alloys that condition their use as thermocouples.

First, these substances should be employed either in a reducing atmosphere (except for tantalum and niobium) or an inert atmosphere, or under a vacuum ($<10^{-3}$ mm Hg), by reason of their high chemical reactivity. This also limits the time that they can be kept in carburated atmospheres. Secondly, the fragility of these materials in the form of wires in a crystalline state, especially in the case of pure tungsten, requires great care and extreme precautions in handling them.

3) Composition of Thermocouples

The relatively easy working of refractory alloys permits producing them either as wires of small diameter (a few hundredths of millimeters as a minimum) or as tubes of small cross section (1 mm \times 0.7 mm) and so making up thermocouples of small size.

Electrical insulation of the wires should be provided by a material possessing particular characteristics throughout the intended range of temperature. It must in fact have a very high resistivity combined with an /20 excellent thermal conductivity, as well as good chemical compatibility with the alloys present. These conditions are generally realized with refractory oxides such as thoria (ThO_2), magnesia (MgO), zirconia (ZrO_2), alumina (Al_2O_3), and glucina (BeO); comparison of their respective merits [19] motivated the choice of the last mentioned for the majority of cases.

On account of these considerations thermocouples are ordinarily made in three forms, cross sections of which are represented diagrammatically in Figure 7.

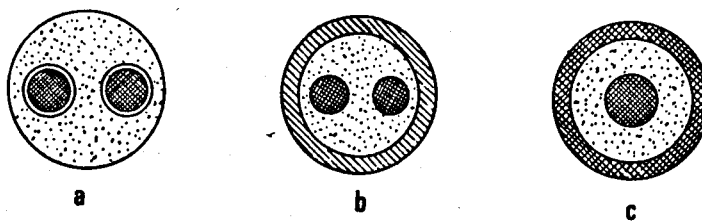


Figure 7

- As bare wires of relatively large diameter (several tenths of millimeters) insulated by bifilar beads of refractory material,
- Under an impervious sheathing of refractory metal (molybdenum, tantalum, or niobium) different from the materials composing the thermocouple itself, the diameter of which is generally from 1 to 2 mm,
- As a coaxial tube (in this case the tube constitutes one of the branches of the thermocouple), permitting still further reduced dimensions.

In the last two cases the electrical insulation is provided by glucina

previously degasified at high temperature and reduced to the state of very closely packed powder. Moreover, the tube or sheathing should be free of any chemical component that might alter the purity of the insulator.

III. THERMOCOUPLES OF TUNGSTEN-RHENIUM ALLOYS

Before taking up the actual study of thermocouples of tungsten-rhenium alloys, let us briefly review the specific characteristics of these materials.

1) Composition

Chemical analysis of the basic metals reveals the presence of impurities, the content of which does not exceed 0.01%, and which are largely molybdenum, iron, aluminum, oxygen, and carbon [13].

The tungsten and rhenium components of the alloys are as follows: /21

<i>Designation</i>	<i>Content (in % by Weight)</i>
W	99.95% tungsten
W 5% Re	5.3 % rhenium
W 26% Re	27.4 % rhenium

2) Diagram of Tungsten-Rhenium Equilibrium

The equilibrium diagram [20] represented by Figure 8 shows a solid phase β limited to 28% rhenium, which includes the alloys studied and which is preserved up to a temperature of about 3000°C.

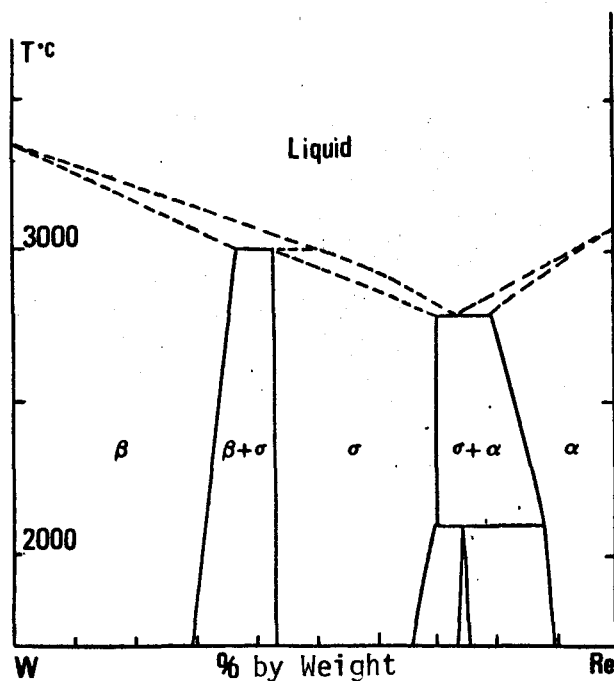


Figure 8. W-Re Equilibrium Diagram.

3) *Physical and Mechanical Properties*

While the properties of pure tungsten may be well known, that is not true of its alloys with rhenium. It is only recently [21,22] that some of these properties have been determined.

It is known, however, that tungsten-rhenium alloys have mechanical properties (breaking strength, elasticity, fragility, etc.) superior to those of pure tungsten, and more markedly so the higher the percentage of rhenium (at least in the phase β). That is why W 5% Re is used instead of pure tungsten to make this type of thermocouple.

The coefficient of diffusion, which in part determines the stability of thermocouples made of alloys, has been measured for tungsten and for the rhenium in the tungsten up to 5200°C [23]. The values determined with the aid of radioactive tracers of these elements are as follows:

$$D (W/W)_{\text{cm}^2/\text{sec}} = 43 \exp (-153/RT)$$

$$D (Re/W)_{\text{cm}^2/\text{sec}} = 275 \exp (-163/RT)$$

For temperatures of about 2600°C these coefficients are accurate in both cases to within $2 \cdot 10^{-10} \text{ cm}^2/\text{sec}$.

4) *Constitution of the Thermocouples Studied*

Since the manufacture of thermocouples of the type of Figure 7c has only very recently been perfected for tungsten-rhenium alloys (sheath W 26% Re, core W 5% Re), only the two types represented by Figures 7a and 7b composed the subject of our study:

- thermocouples of bare wires of diameters 0.3 mm, 0.5 mm, and 0.8 mm, the 22 welding of which is done by mercury arc,
- thermocouples sheathed in tantalum or niobium, the dimensions of which are as follows:

<i>External Diameter of Sheath</i>	<i>Internal Diameter of Sheath</i>	<i>Diameter of Wire</i>	<i>Maximum Length</i>
1.8 mm	1.5 mm	0.30 mm	1 m
1.3 mm	1.0 mm	0.20 mm	1 m

The welding of the wires and if necessary the closing of the sheath in the hot part are done by arc welding under argon.

STUDY OF TUNGSTEN-RHENIUM ALLOY THERMOCOUPLES
OUTSIDE THE REACTOR

A. PRELIMINARY STUDIES

I. EXPERIMENTAL APPARATUS

The general principle of the method used in studying the thermocouples is as follows [30]:

a) Production of the high temperatures. -- The thermocouple is placed in a refractory container which may be equated with a black body, situated inside a furnace attaining temperatures of about 2300°C in a molecular vacuum.

b) Measurement of the temperatures. -- This is provided

- between 0° and 1400°C by a standard Pt/Pt 10% Rh thermocouple,
- between 1050°C and 2300°C by optical pyrometry,
- by fixed points (solidification of pure elements) throughout the temperature range.

c) Measurement of the electromotive force. -- The opposition method is used, either with a recording instrument (MECI, Speedomax type with several tracks) or by means of a millivoltmeter-potentiometer (MECI, type ESPM).

1) Description and Characteristics of Furnace I

The furnace described here, a cut-away view of which is given in Figure 9, was chiefly used for the statistical study of thermocouples and of their stability in time.

a) The heating element (Figure 10). -- The heating element is made of tungsten wire (diameter 5 mm) coiled in a double spiral in order to facilitate expansion and avoid field effects.

The usable inside volume is of the following dimensions: diameter 45 mm, height 85 mm. Current is supplied through molybdenum rods (diameter 12 mm) which slide in through airtight openings and are cooled by a water circulation system.

b) The crucible. -- Inside the resistor is placed the isothermal cell, consisting of a graphite crucible held in place by a tantalum plate. A lateral opening (diameter 4 mm) permits observation of the radiation emitted from this black body.

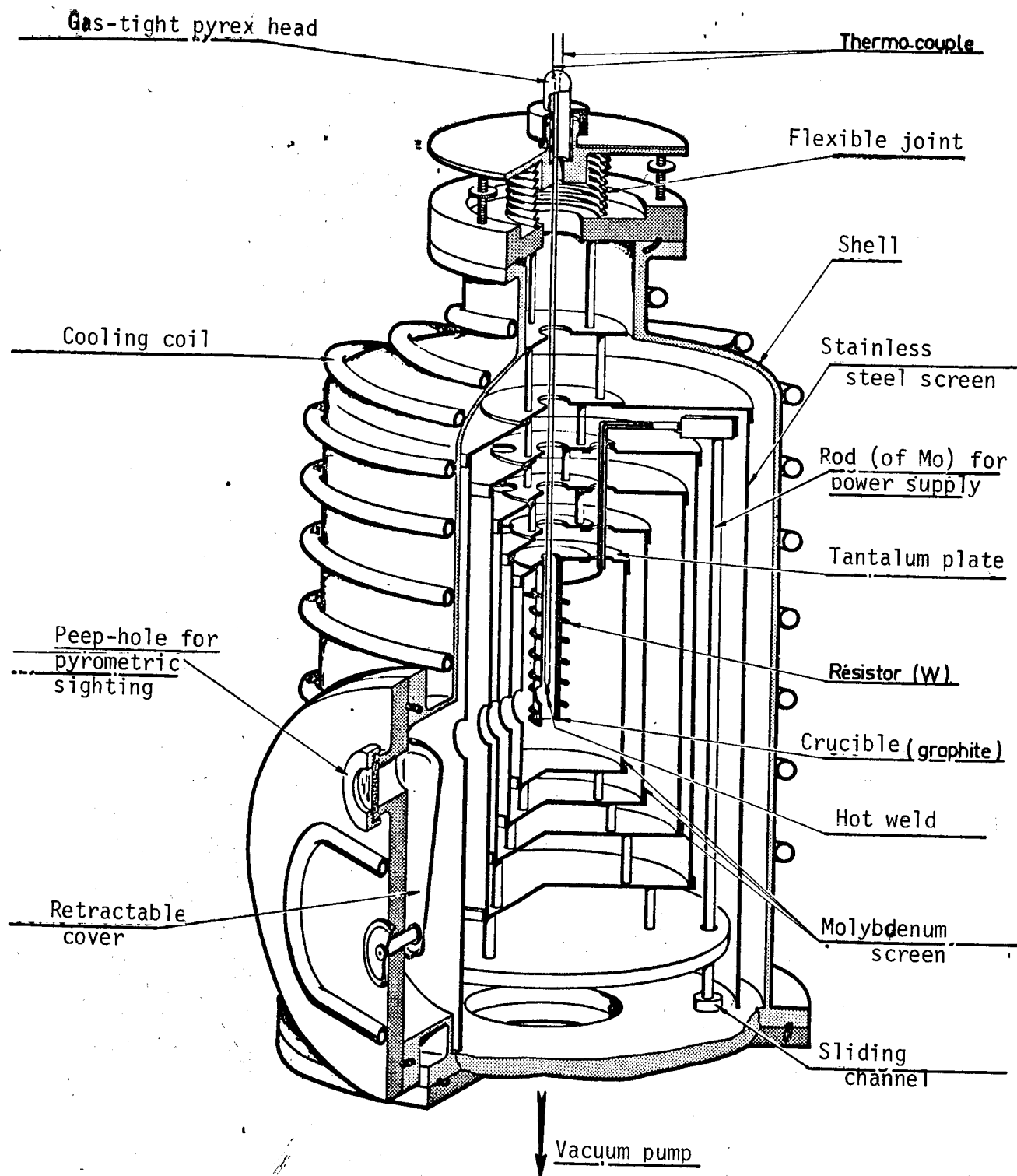


Figure 9. Cut-away View of Furnace I.

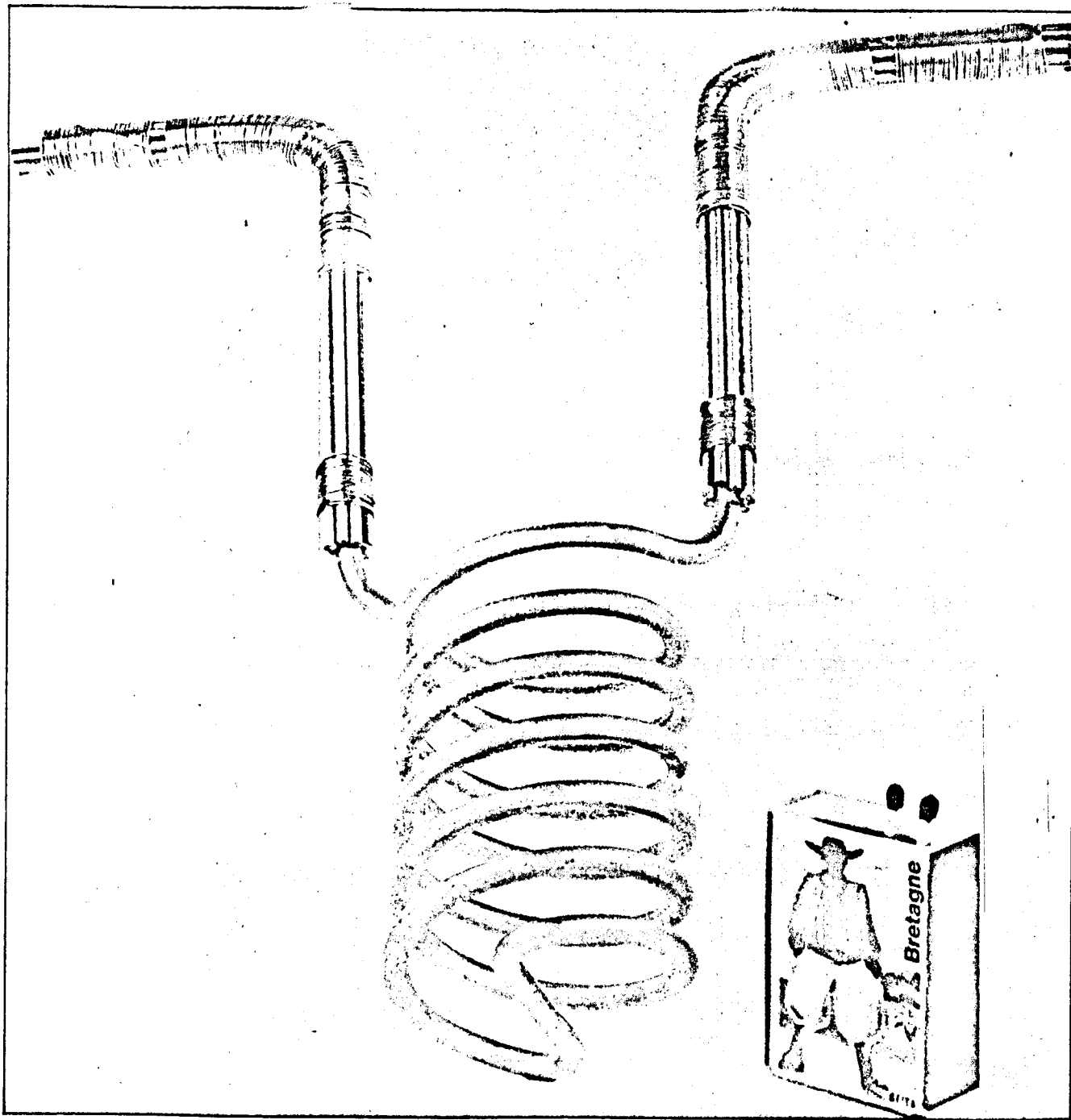


Figure 10. Heating Element of Furnace I.

c) *The thermal screens.* -- The heating element is surrounded by four cylindrical molybdenum screens enclosed in a stainless steel screen.

In operation the temperature of the first screen will assume the same value as that of the crucible; to guard against mechanical deformations, four tungsten bands riveted lengthwise ensure its rigidity.

At the top and bottom, thermal insulation is obtained by means of circular molybdenum plates with recessed edges into which the lateral screens are set.

These screens are supported by a copper plate cooled internally by circulating water.

d) *The outer shell.* -- The components just described are enclosed in a steel shell cooled by circulating water.

Optical pyrometric observations are done through a "Pyrex" glass peephole. To prevent soiling its surface, a retractable cover which is removed only for the time necessary for sighting has been mounted inside the shell.

e) *The pumping unit.* -- The furnace is installed on a pump bench provided with a primary blade-type pump and an oil diffusion pump which makes it possible to obtain an inside pressure of 10^{-3} mm Hg even during high-temperature degasifications.

f) *Electric power supply.* -- An autotransformer followed by a voltage-reducing transformer supplies the resistor. The flexibility and precision of such an apparatus ensure sufficient stability to achieve thermal equilibrium at any temperature. /29

g) *Electrical and thermal characteristics of the furnace.* -- Measuring the voltage between the chilled channels which constitute the current intake and taking the intensity that crosses the resistor have enabled us (Figure 11) to translate the variations of the power consumed by the furnace as a function of the temperature of the isothermal cell. It may be noted that at 2350°C the power of 15 kVA corresponds to an intensity of 500 amperes.

The curve shown in Figure 12 illustrating the development of the temperature of the cell as a function of the time after the current is cut off brings out the thermal inertia of the furnace.

2) *Installation of the Thermocouple*

It is indispensable to be able to introduce into the furnace the thermocouple to be studied, and sometimes also the reference thermocouple, with a minimum dislocation of components. To this end the thermocouple, insulated by refractory beads, is first mounted on a glass tube which has been made gas-tight and fixed by means of metal rings and neoprene joints to a piece mounted at the top of the outer shell.

A flexible tombac membrane provides for the vertical and lateral motion of the thermocouple. This arrangement also enabled us to establish the excellent homogeneity of the temperature inside the crucible.

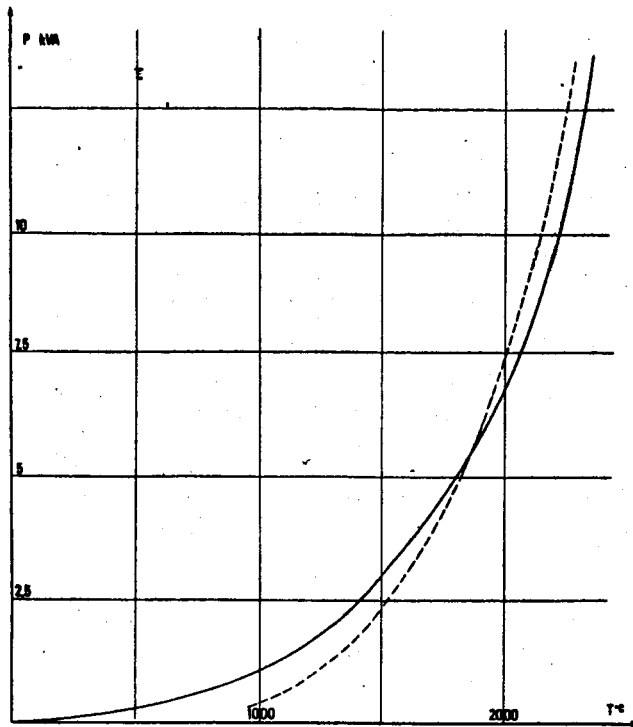


Figure 11. Actual Power Consumed and Theoretical Power Emitted by Radiation. /30

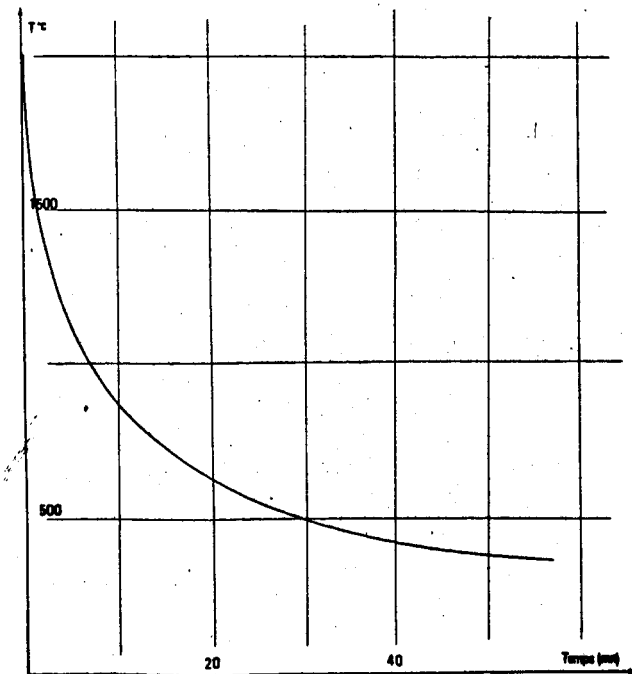


Figure 12. Cooling Curve.

In consequence of their design, sheathed thermocouples require a special device for their installation, diagrammed in Figure 13.

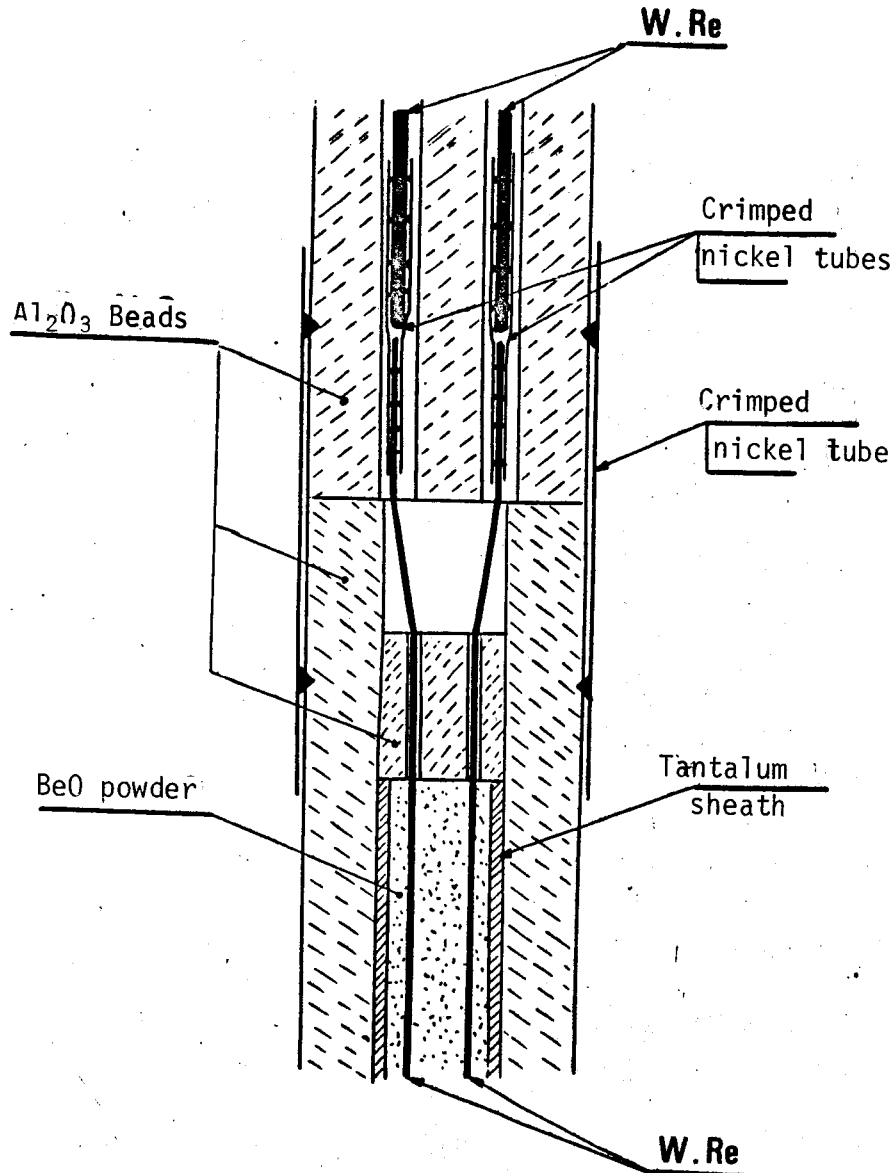


Figure 13. Apparatus for Installing Sheathed Thermocouples.

3) *Measurement of the Electromotive Force -- Precision*

On the outside of the shell the wires of the thermocouples are fixed to two blocks of copper embedded in the molten glass by way of test tubes containing vaseline oil. Copper cords connect them to the measuring apparatus.

The recording device claims an accuracy of $\pm 15 \mu\text{v}$ in the ranges used;

The ESPM affords a measurement subject to an absolute error of $\pm 8 \mu\text{v}$.

4) Measurement of the Temperature -- Precision

/32

The cold weld device makes it possible to obtain a reference temperature of between 0° and 0.5°C.

The Pt/Pt 10% Rh thermocouple installed simultaneously with and in the same manner as the thermocouple to be studied affords a precision of about $\pm 1^\circ\text{C}$.

Between 1050° and 2300°C the temperature is measured by means of a micro-pyrometer with disappearing filament (Jobin-Yvon) through the viewing window, the absorption of which has been previously determined.

The accuracy of the micropyrometer is as follows:

$$\Delta T_{1050^\circ\text{C}} = \pm 3^\circ\text{C}$$

$$\Delta T_{2000^\circ\text{C}} = \pm 7^\circ\text{C}$$

In fact the crucible behaves strictly as a black body only above 1500°C. This phenomenon, shown by comparison of measurements on the standard thermocouple and on the micropyrometer, indicates a disparity between the two methods of 25°C at 1050°C, which diminishes with the rising temperature and becomes zero above 1500°C.

To eliminate this indeterminateness we were led to employ the method of fixed points, which, in agreement with the measurement with the standard thermocouple, allowed us to correct the pyrometric reading.

The principal fixed points of the thermometric scale were thus utilized. The fusion of a wire of the metal under consideration on the cold weld or the immersion of the latter in a bath of the melted substance are shown by an emf bearing on the recording of the temperature as a function of time.

II. STATISTICAL CALIBRATIONS; DISPERSION; MEAN CALIBRATION CURVES

We proceeded to calibrate a large number of thermocouples (an average of 20 for each type) made of W/W 26% Re and W 5% Re/W 26% Re, in order first to determine the maximum dispersion due to variations in composition among the various manufacturing batches and secondly to establish a mean calibration curve for each category and shape of thermocouple [30].

1) Calibration Conditions

The emf's delivered by the thermocouples are generally read every 50°C, after complete stabilization of the temperature (5 minutes on the recording).

After calibration the thermocouples are subjected to a preheating at about 2000°C for 15 minutes. The recrystallization occasioned by this treatment avoids the variable effect of the initial cold working. Whatever the nature of the thermocouple, it is well known that a preliminary heating is always recommended.

/33

2) Statistical Calibrations; Spread

For each type of thermocouple we have plotted on one and the same graph the set of points obtained by calibration. The zone within which the points are included enabled us to deduce the maximum deviation $\pm\Delta T$ with respect to the mean value.

Figure 14 indicates as a function of the temperature the variations of the absolute value $|\Delta T|$ of this deviation and of the corresponding relative value $\frac{\Delta T}{T}$.

Note 1: These results are favorable for the combination W/W 26% Re. We believe that the slightly wider spread with W 5% Re/W 26% Re thermocouples is due to the greater influence of variations in composition from one batch to another of the W 5% Re alloy.

Note 2: We never found any systematic deviation due either to the diameter of the wires in the case of bare-wire thermocouples or to the material constituting the sheath (tantalum or niobium) for sheathed thermocouples.

3) Calibration Curves

In order to draw definitive calibration curves for each type of thermocouple, we took at a given temperature the mean value of the emf in the zone of spread.

It should be noted that the errors arising from the measuring instruments are far slighter than the spread of purely thermoelectric origin.

We thus established mean calibration tables correct to two decimal places (0.005 mv). This accuracy permits differential measurements to:

$\pm 0.5^{\circ}\text{C}$ between 700° and 1700°C } in the case of W/W 26% Re,
 $\pm 0.7^{\circ}\text{C}$ at 2000°C

and to

$\pm 0.5^{\circ}\text{C}$ between 400° and 1300°C } in the case of W 5% Re/W 26% Re.
 $\pm 0.8^{\circ}\text{C}$ at 2000°C

/35

The mean calibration curves and tables for the thermocouples are assembled at the end of this treatise.

4) Characteristics of the Thermocouples

a) *Bare-wire W/W 26% Re thermocouples.* -- Their excellent linearity is reflected in a sensitivity of $21 \mu\text{V}/^{\circ}\text{C}$ between 700° and 1700°C . By 2000°C this has decreased but is still near $14.5 \mu\text{V}/^{\circ}\text{C}$.

This type of thermocouple, while it has very good thermoelectric characteristics, has the disadvantage of being excessively fragile after recrystallization in consequence of the presence of pure tungsten.

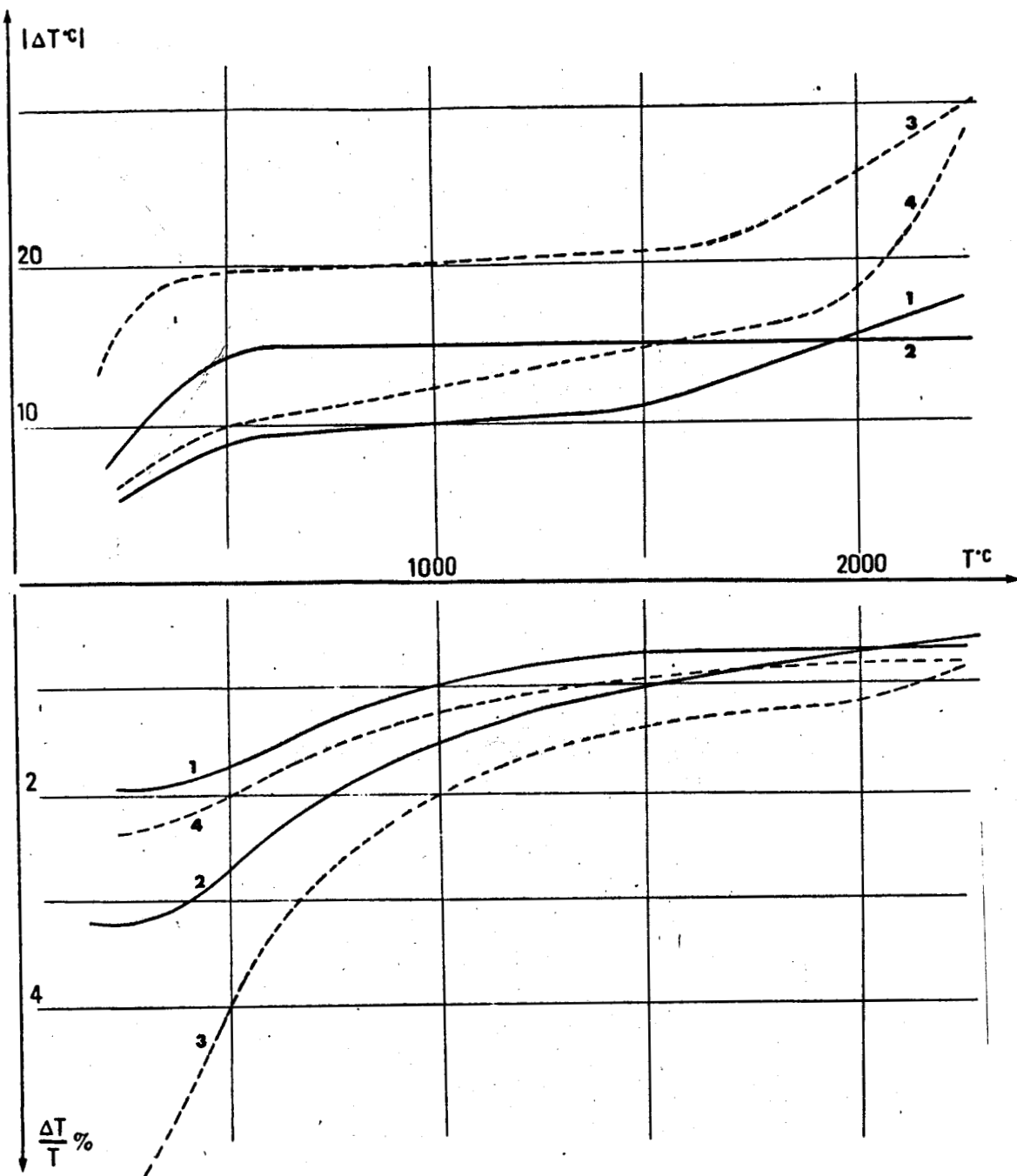


Figure 14.

- | | |
|--------------------------------|--------------------------------------|
| 1) W/W 26% Re, Bare Wires | 3) W 5% Re/W 26% Re, Bare Wires |
| 2) W/W 26% Re, Sheathed 1.8 mm | 4) W 5% Re/W 26% Re, Sheathed 1.8 mm |

b) Bare-wire W 5% Re/W 26% Re thermocouples. -- Between 400° and 1300°C their thermoelectric power is practically constant and is equal to 18.5 $\mu\text{V}/^{\circ}\text{C}$, decreasing then to 12 $\mu\text{V}/^{\circ}\text{C}$ in the neighborhood of 2000°C.

c) *Sheathed thermocouples.* -- Their thermoelectric characteristics are essentially identical to those of bare-wire thermocouples, although they exhibit an appreciable deviation from the latter at high temperature depending on the dimensions of the sheath; thus the sheathed W/W 26% Re thermocouple (external diameter 1.8 mm) delivers a higher emf at 2000°C by 1 mv than the same thermocouple made up with bare wires.

Various hypotheses may be advanced to explain this anomaly. In particular it is conjectured that a diminution of the resistivity of the glucina at high temperature induces the appearance of a contact emf between the sheath and the wires which disturbs the measurements.

We have not established the *cal* calibration curves for sheathed thermocouples of external diameter of 1.3 mm. In fact the number of specimens calibrated was insufficient for the purposes of a statistical study. Nevertheless, the few measurements made showed a result identical to that of the sheathed thermocouples of 1.8 mm external diameter.

d) *Comparison with thermocouples of various provenience.* -- /37
Figure 15 gives the curves $E_{mv} = f(T^{\circ}C)$ for thermocouples of various origins -- Bocuze, Hoskins, Enghelart [*sic*; not in the figure], Thermo-Electric, which we calibrated or for which we accepted the calibration curve furnished by the manufacturer.

The differences found are essentially due to variations in composition from one method of manufacture to another.

Note: In the derivations given below we shall frequently have occasion to introduce the quantity ΔT which represents a difference in temperature between the initial calibration and the calibration done after the thermocouple has been subjected to the influence of some phenomenon or other.

We have adopted the convention of marking ΔT with the sign (+) if for a given value of the emf the evolution is reflected by a rise in the temperature indicated, and with the sign (-) in the contrary case.

III. STABILITY IN THE COURSE OF TIME

In order to determine any variations of the emf delivered by thermocouples subjected to long periods of heating, various types were left uninterruptedly at 1500°C for 150 hours under a secondary vacuum.

These data imposed by the characteristics of the furnace (which if it were kept at a higher temperature for a longer period of time might undergo damaging overheating) are nevertheless comparable to the conditions of industrial use and are adequate to give an idea of the behavior of the thermocouples.

The tests done on thermocouples previously subjected to stabilization heat treatment and then calibrated did not modify discernibly their thermoelectric characteristics. In fact the differences observed between calibrations before and after prolonged heating under the stated conditions are of the order of magnitude of errors attributable to the experimental apparatus (cold welding, measurement of the emf, etc.).

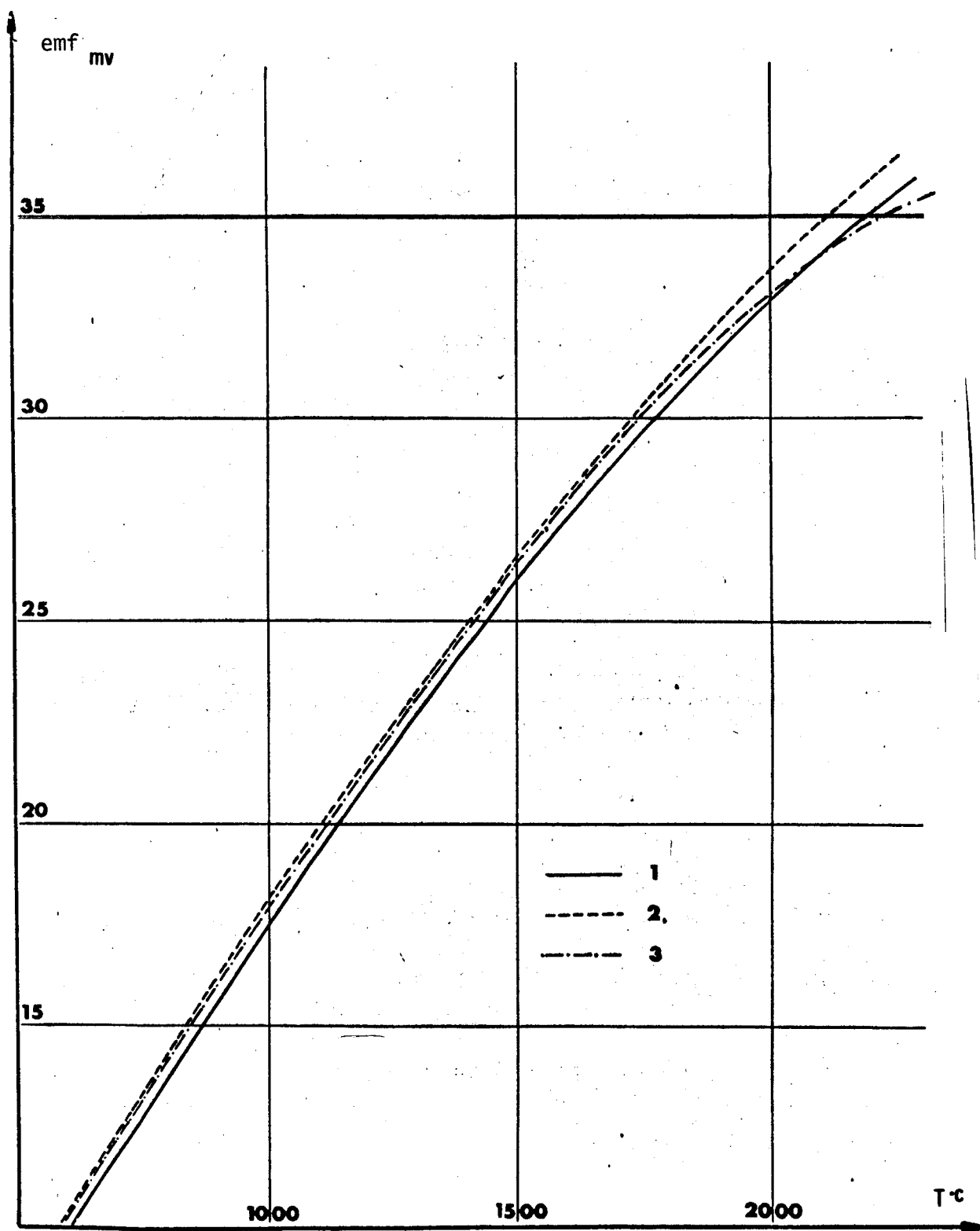


Figure 15. Sheathed W 5% Re/W 26% Re Thermocouples.

1) Bocuze; 2) Hoskins; 3) Thermo-Electric

These results, which are in accord with the work of Lachman [29], find their justification in the following facts:

1) The low values of the coefficients of diffusion of tungsten and rhenium do not permit great variation in the composition of alloys in the vicinity of hot welding. /38

2) The physical properties of these alloys, recrystallized after stabilization heating, are not necessarily modified by a prolonged heating which does not involve thermal shocks capable of causing further metallurgical transformations.

IV. STABILITY IN GASES

All our operations were carried out under a secondary vacuum. Indeed, the design of the apparatus does not allow of attaining high temperatures in the presence of inert gases or reducing gases, since their thermal conduction occasions a distinct increase in caloric exchanges and entails a significant rise in the electric power consumed.

For that reason it seems to us of interest to review briefly the conclusions reached by Lachman [29] on the basis of his experiments on tungsten-rhenium alloy thermocouples under a gaseous atmosphere, largely hydrogen and argon.

1) Influence of Hydrogen

His work led to the conclusion that these thermocouples are remarkably stable under hydrogen (especially for the type W/W 26% Re), particularly after a stabilization treatment (preliminary heating), and that there is no contra-indication to their use in the presence of that gas.

2) Influence of Argon

Figure 16 shows the results of tests of stability in time on W/W 26% Re thermocouples heated to 1000° and 2200°C under an argon stream (150 l/hour).

The variations thus observed are lower than the spread of strictly thermoelectric origin observed by Lachman in his samples. It follows that the use of such thermocouples under argon is entirely satisfactory.

Note: No systematic studies under helium have yet been carried out. The conclusions relative to the tests described above lead us to think that the presence of helium would introduce only very slight modifications in the characteristics of these thermocouples. /39

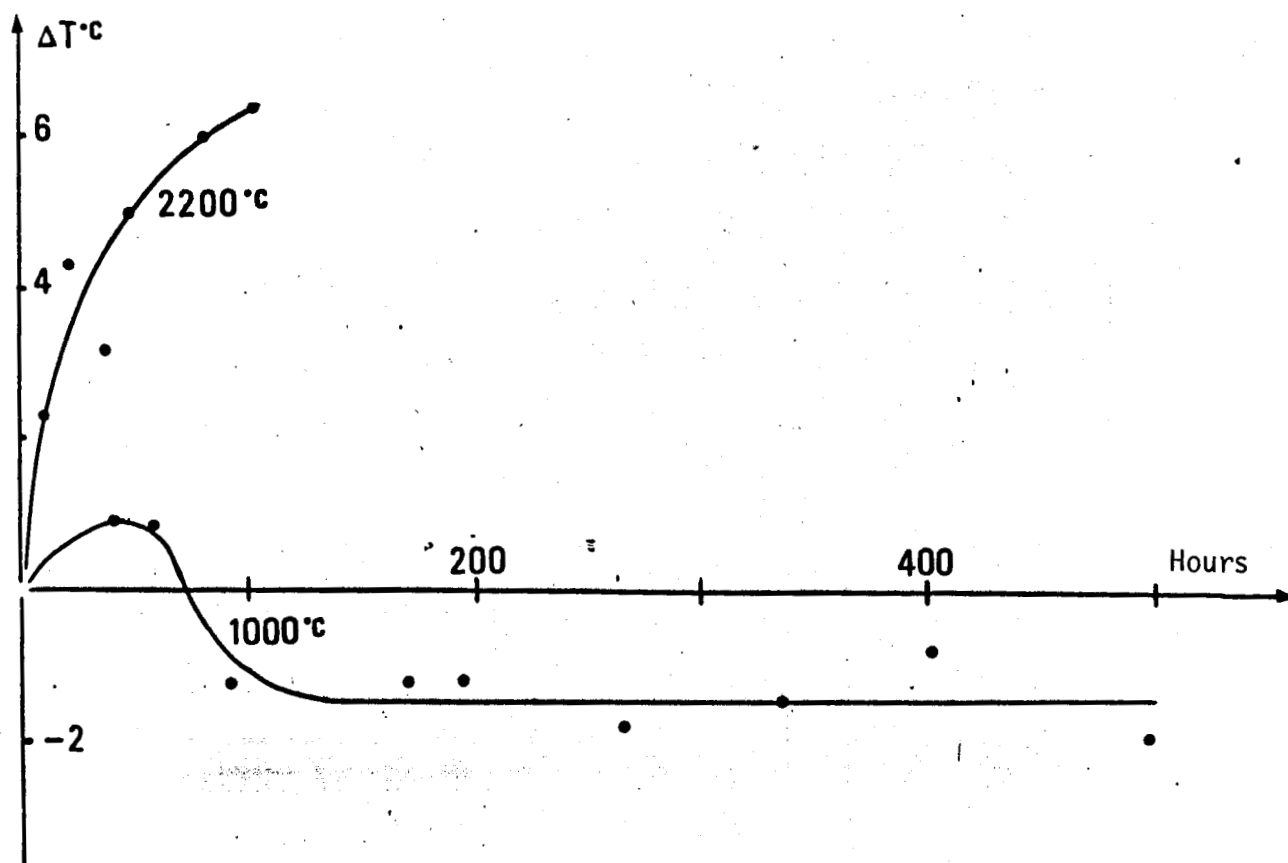


Figure 16. Stability of W/W 26% Re Under Argon [29].

Study of the influence of thermal shocks on the behavior of thermocouples and measurement of their response time necessitated construction of a furnace (II) which had largely the same characteristics as that described above but which could be fitted with apparatus making it possible to do the tests under consideration.

I. EXPERIMENTAL APPARATUS

1. Description and Characteristics of Furnace II

Furnace II, the operating principle of which is identical to that of Furnace I, is shown in Figure 17, where the various detail modifications incorporated in it can also be seen.

a) The Heating Element. -- In order to improve the resistance of the heating element, we used thoriated tungsten (0.4% Th) in the place of pure tungsten.

b) The Outer Envelope. -- Made up of a double stainless steel wall equipped with baffles on the inside to ensure a more efficient cooling, its upper part is covered by a plate which is also cooled and on which auxiliary apparatus can be mounted.

Laterally a sighting window permits pyrometric observations of five different points. In fact this furnace was also designed for possible study of the influence of thermal gradients along the length of thermocouples.

2. Thermal Shock Apparatus

/42

a) Principle. -- This apparatus, a cut-away view of which is presented in Figure 18, provides for moving the thermocouple at various speeds in such a way that its hot weld can pass from a hot zone to a cold zone or vice versa, the furnace being kept at constant temperature under a secondary vacuum.

Moreover the five speeds provided (varying in the ratios $\frac{1}{2}$, 1, 5, 10, 20) provide for maximum displacement within the period of 8 minutes and 15 seconds.

II. INFLUENCE OF THERMAL SHOCKS

We will define "a thermal shock" as being the successive rise and fall of the temperature of the hot weld of the thermocouple, or vice versa, without gradations, and with identical rates of heating and cooling calculated as functions of the difference ($T_{\max} - T_{\min}$) between the extremes of temperature attained.

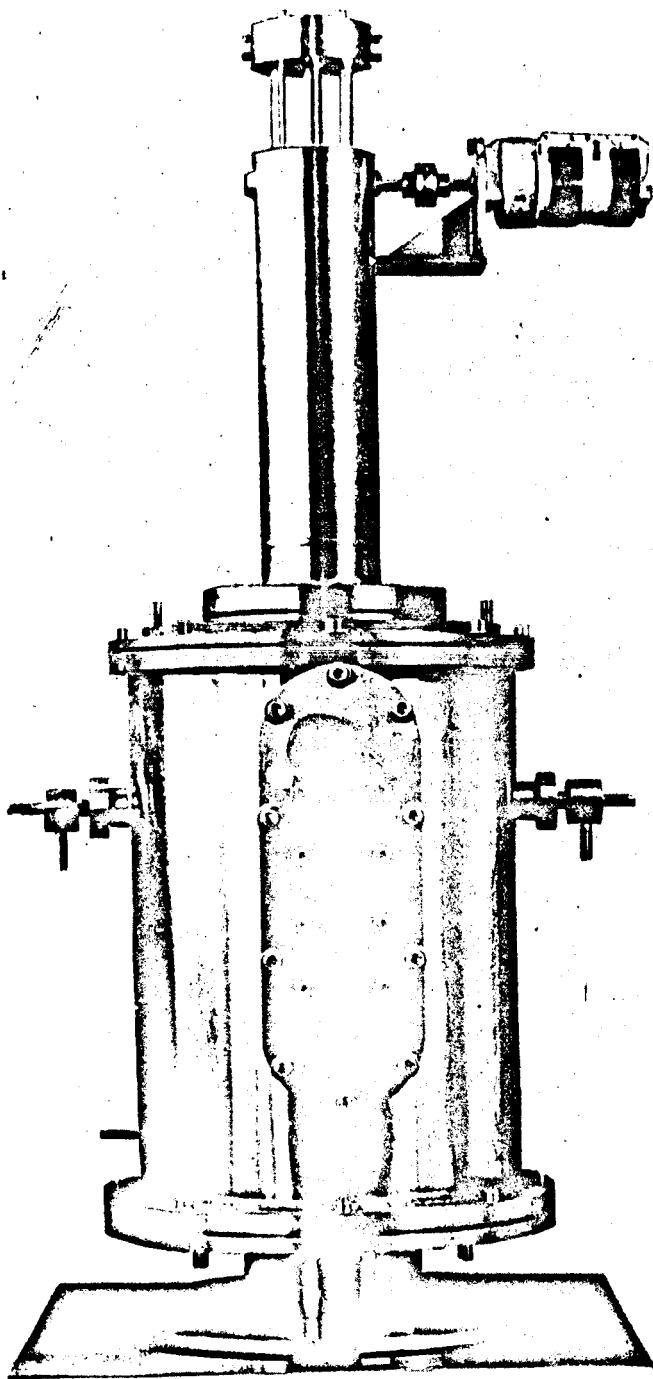


Figure 17. Furnace II with Thermal Shock Apparatus.

1. Preliminary Tests

a) Experimental Method.

-- The first tests were done on Furnace II, by rapidly (or progressively) raising the temperature and by suddenly (or ing off (or slowly reducing) the power supply.

The mean rates

$$\frac{T_{\max} - T_{\min}}{\Delta t}$$

of cooling and heating thus obtained do not exceed 5°C per second.

b) Results. -- The studies done on thermocouples of various types yielded the following results:

- Between 2000°C and /45 1000°C, for rates lower than 1°C per second, no modification of the thermoelectric characteristics of the thermocouples was observed, regardless of the number of thermal shocks.

- On the other hand, in the same temperature range but with rates on the order of 5°C per second, after three or four thermal shocks a considerable change shows up in the emf delivered. Figure 20 shows the differences ΔT between calibrations before and after the thermal shocks to which a thermocouple was subjected consisting of bare wires (0.8 mm in diameter) of W 5% Re/W 26% Re. Identical results were obtained with sheathed thermocouples.

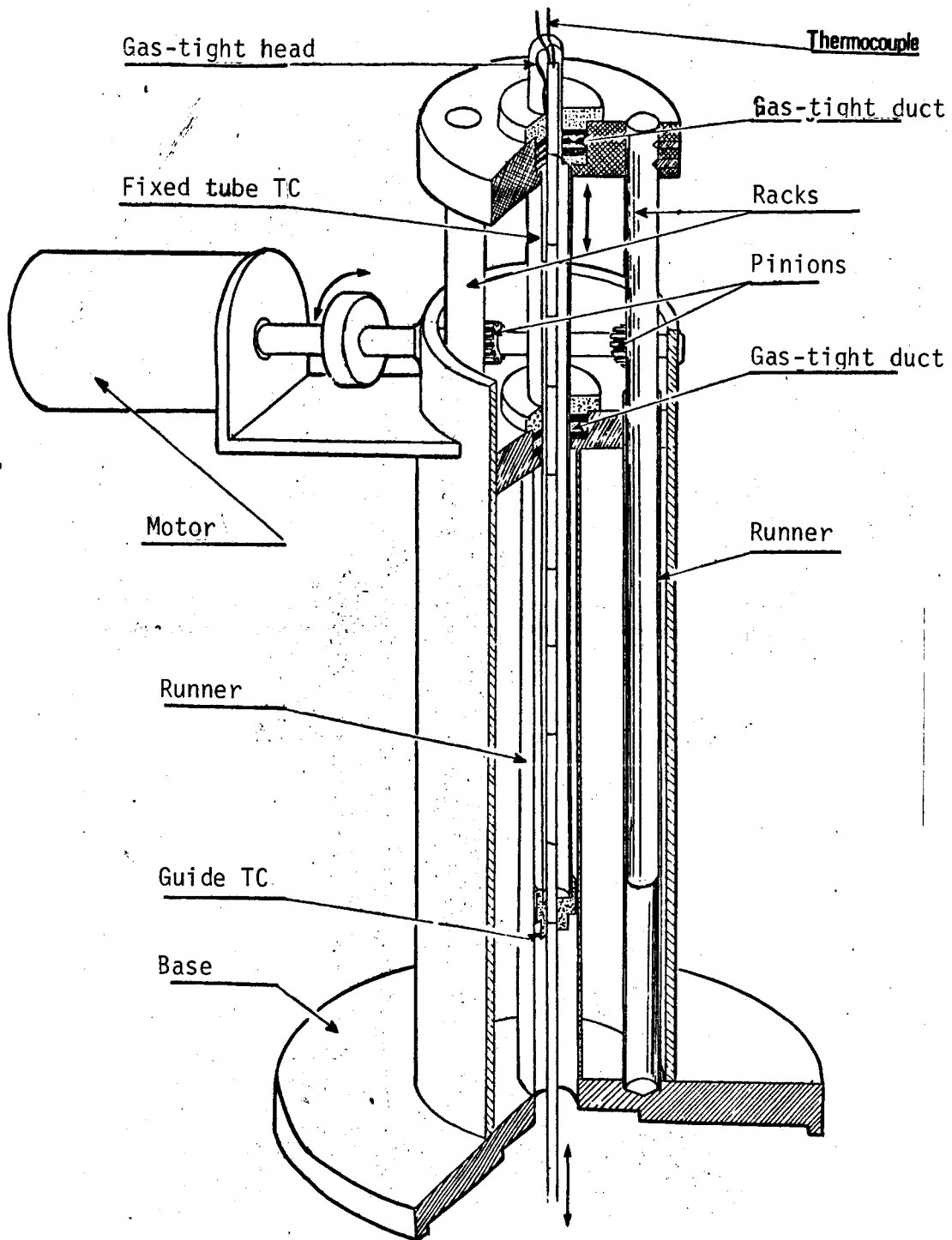


Figure 18. Thermal Shock Apparatus.

2. Tests Done with the Thermal Shock Apparatus

a) *Experimental Conditions.* -- We carried out our experiments with the temperature T_{\max} set at 2000°C . This means in the present case the temperature at which the crucible is kept during the operation. Under these conditions it turns out that the temperature of the cold zone is equal to 250°C .

Taking into account these extremes of temperature (determined by the prior measurement of the thermal gradient along the course traveled by the thermocouple) and the five displacement speeds at our disposal, the curves $T^{\circ}\text{C} = f(\text{time})$ of Figure 19 represent as a function of the time the temperature of the region in which the hot weld of the thermocouple is situated during its translation.

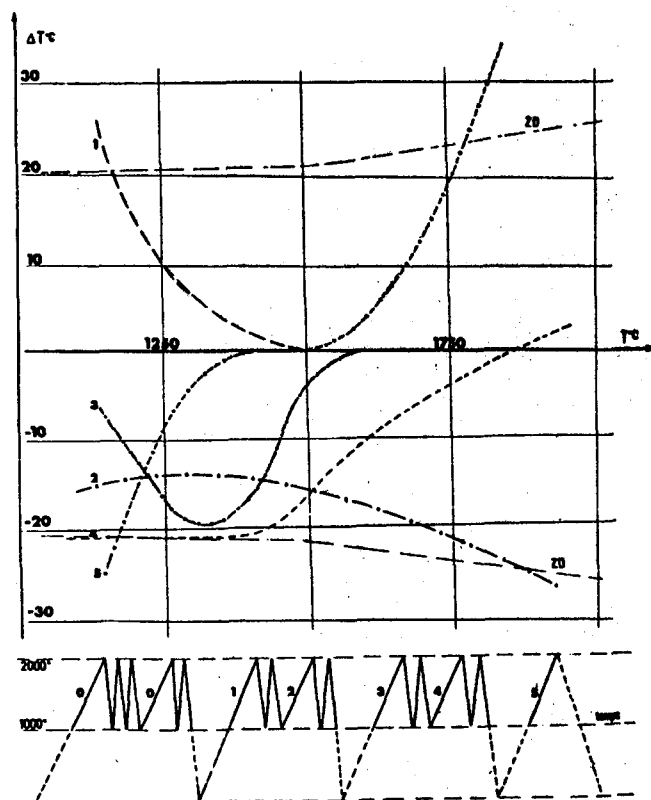


Figure 19. Temperature Gradient.

We assigned to each curve a mean velocity (rate) $V_m(T)$ defined as above and a maximum velocity $V_M(T)$ corresponding to the linear part of those curves:

$$V_M(T) = \frac{1750^{\circ} - 250^{\circ}}{\Delta t}$$

In fact the thermal gradient is linear only between 1750° and 250°C .

The following table gives the values of these speeds for the different curves considered.

	V (T) $^{\circ}\text{C}/\text{sec}$	V (T) $^{\circ}\text{C}/\text{sec}$
I	3.5	5.5
II	6.5	10
III	30	50
IV	60	90
V	115	185

b) *Results.* -- By way of example, Figures 20b and 20c show the deviations ΔT (defined on page 23) induced by thermal shocks of a given rate (velocity) on two of the thermocouples studied.

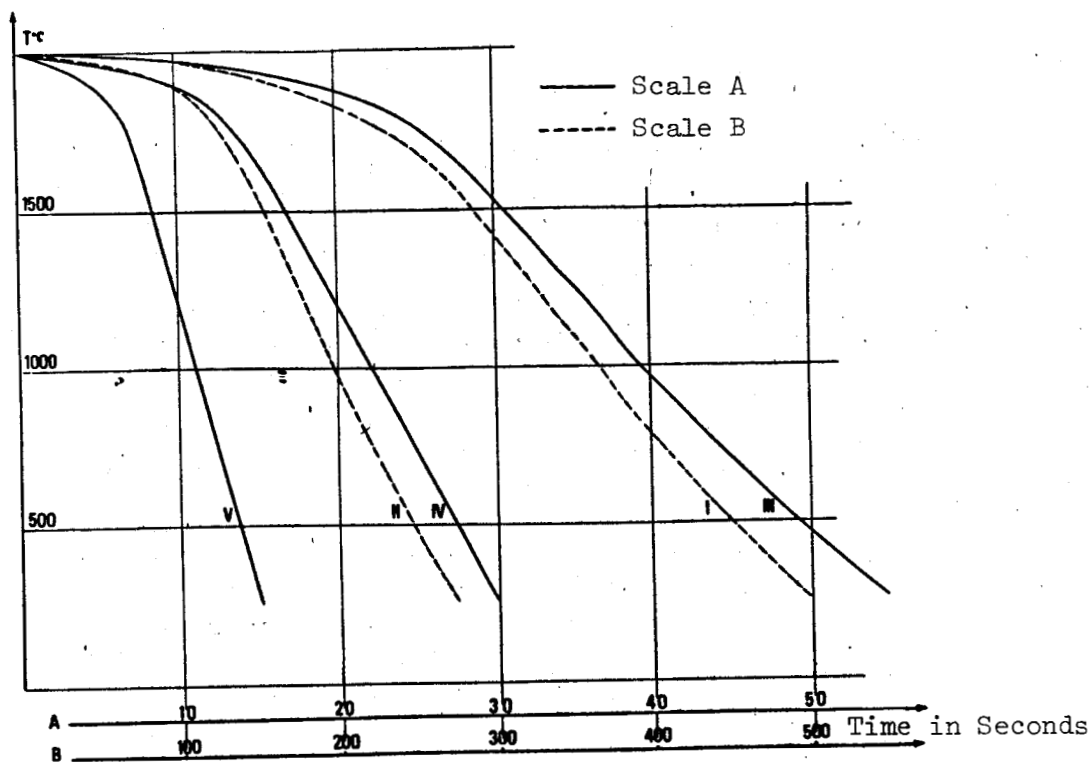


Figure 20a. W 5% Re/W 26% Re. Bare Wires, Diameter 0.8 mm.

3. Conclusions

a) *Influence of the Rate and Number of Thermal Shocks.* -- The experiments we carried out can be summarized in the following table.

$V_m(T)$ $^{\circ}\text{C}/\text{sec}$ \ Number	Number	
	<4	>4
<3.5	No Influence	
=3.5	No Influence	Deviation
>3.5	Deviation	

b) *Magnitude of the Deviations.* -- On the curves $\Delta T = f(T^{\circ}\text{C})$ represented by Figures 20b and 20c we have drawn in the area of spread of purely thermoelectric origin associated with the type of thermocouple studied. This enables us to state that the deviations affecting thermocouples subjected to

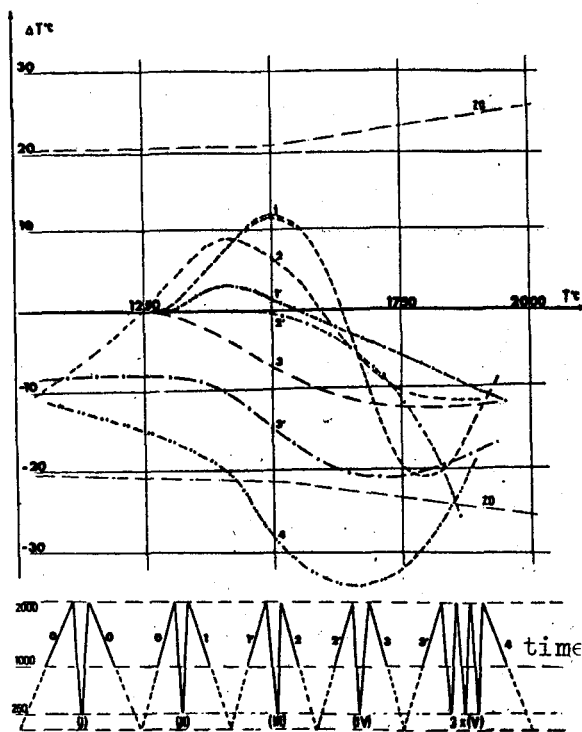


Figure 20b. W 5% Re/W 26% Re
Bare Wires, Diameter 0.5 mm

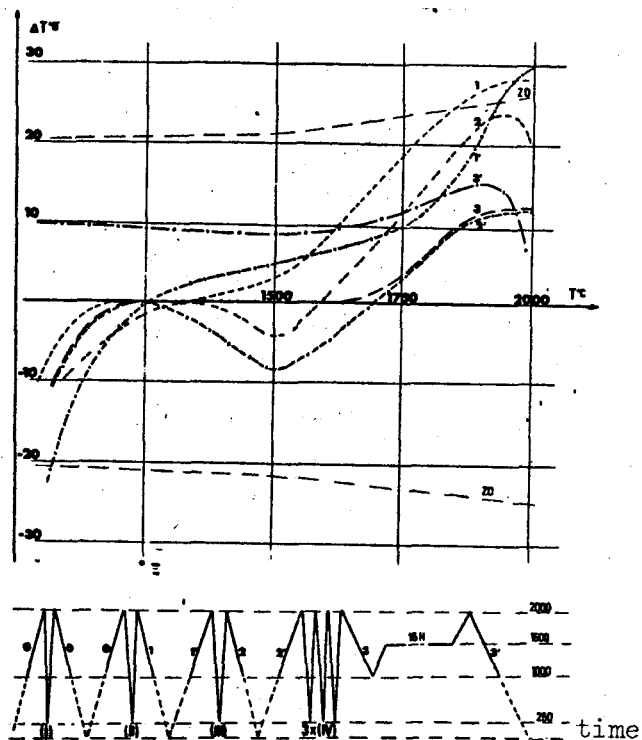


Figure 20c. W 5% Re/W 26% Re
Sheath: Ta, Diameter 1.3 mm

thermal shocks are of the order of magnitude of that spread.

c) *Reproducibility.* -- These curves do not seem to obey a fixed law. In fact, the deviations measured develop in an unpredictable and incoherent way as a function of the number and rate of the shocks. Moreover, a difference appears between calibrations of the type E_n and those of the type E'_n .

d) *Regeneration and Interpretation.* -- Prolonged heating at 1500°C of a thermocouple previously subjected to a stabilization annealing and which has undergone a series of thermal shocks (Figure 20c) does not ensure regeneration, since the deviation is still not eliminated. It thus appears that we are faced here with irreversible phenomena whose variable influence ^{/49} on the emf delivered reflects an unstable state of metallurgical equilibrium and which finds its explanation in the fact that the not entirely crystallized parts are continuously modified by rapid changes in temperature, with resulting changes in grain size, grain orientation, etc.

It would be interesting, in order to confirm this hypothesis, to be able to crystallize the thermocouple (after a previous stabilization annealing) completely in an apparatus designed for that purpose and to subject it to a series of thermal shocks of the same type as those described above.

III. MEASUREMENT OF THE RESPONSE TIME

1. Principle of Measurement

For a temperature range $T_1 - T_0$ in a given environment the response time τ_0 of a thermocouple is the time required by the thermocouple to measure the temperature T_0 when it has been moved from the temperature T_1 to the temperature T_0 at infinite speed.

As we cannot attain an infinite speed of variation experimentally, the response time is defined as being the limit of $\tau = t_2 - t_1$ as $t_1 - t_0$ approaches zero (Figure 21).

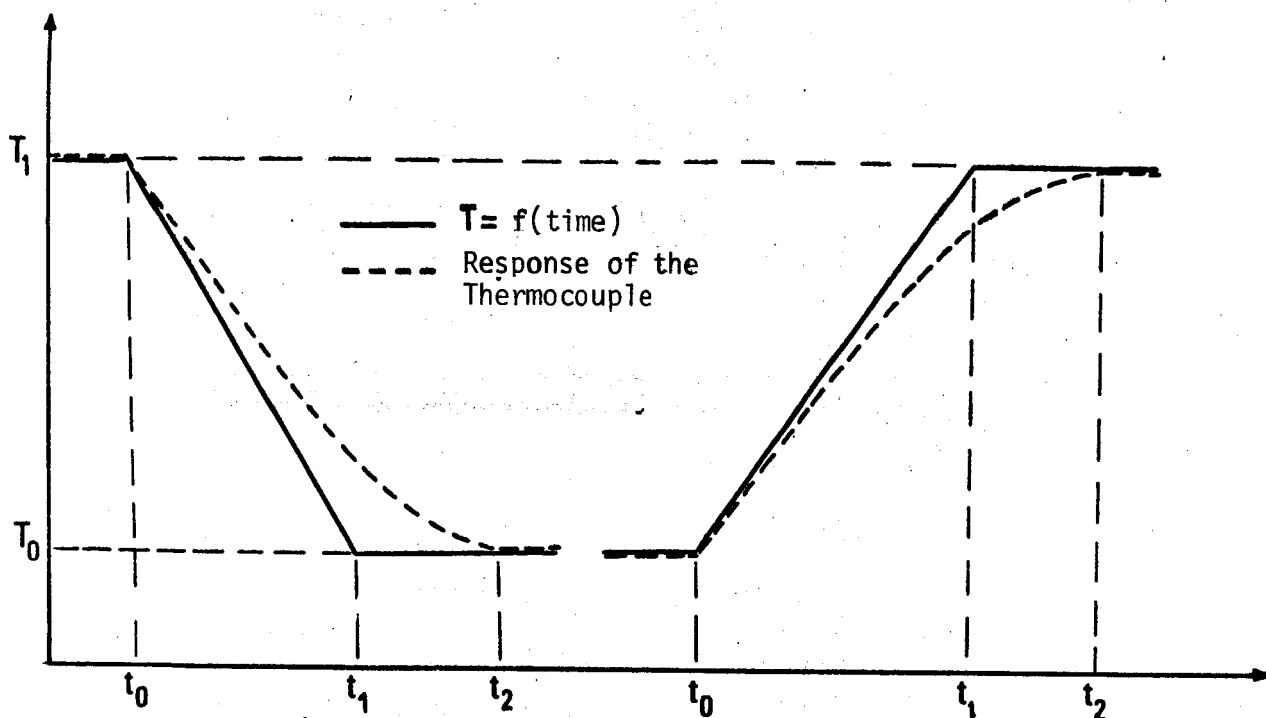


Figure 21.

Let $T = V_T \cdot t$ be the function representing this variation (assumed to be linear) in the range $t_1 - t_0$, where V_T is the speed of variation. Between the limit temperatures in question, we have:

$$T_1 - T_0 = V_T(t_1 - t_0) ; \quad \frac{1}{V_M} = \frac{t_1 - t_0}{T_1 - T_0} .$$

If we measure $\tau = t_2 - t_1$ for several values of V_T and construct the curve $\tau = f(\frac{1}{V_T})$ schematized in Figure 22, extrapolation of that curve to $\frac{1}{V_T} = 0$ yields the value τ_0 sought.

Note: This definition of the response time, which differs from that common-

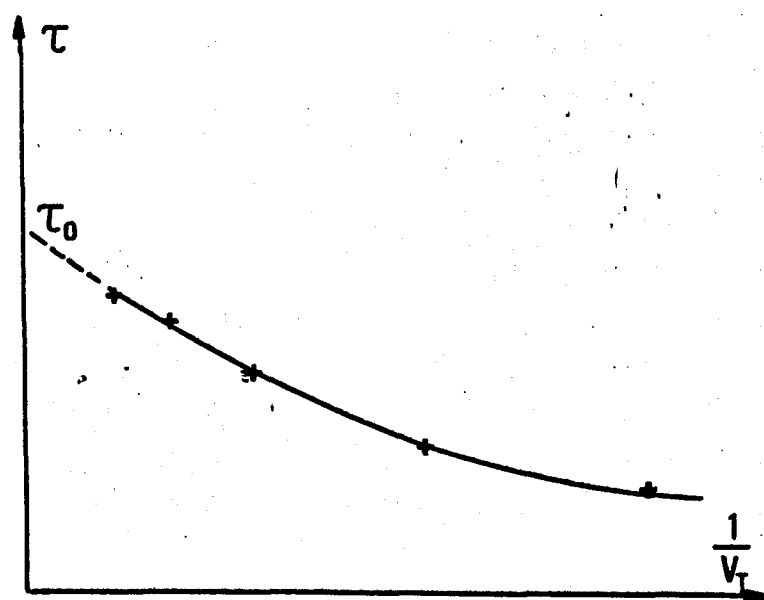


Figure 22.

ly accepted (time required by the thermocouple to measure 63.2% of the difference in temperature to which it is suddenly exposed), corresponds to the real time, and is imposed upon us by the fact that the response curves of the thermocouple obtained experimentally to heating and cooling are not simple exponential curves.

2. Experimental Method

/51

Simultaneously with the thermal shock tests we proceeded to the determination of the response time, as the experimental conditions practically correspond to the principle of measurement. However, as we indicated in the preceding chapter, the speed of variation of temperature is constant only between 1750° and 250°C, for a maximum temperature of 2000°C. It is therefore necessary to take account, in the results obtained, of the curves $T = f(\text{time})$ shown in Figure 19.

3. Results

a) *Time of Response to Cooling.* -- The difficulty of locating the stabilization point (asymptotic curves) forced us to measure the response time at different speeds at a temperature 10°C higher than the temperature of the cold zone.

In cooling, as the rates of variation of the temperature are constant starting at 1750°C (conditions approximating the principle of measurement), we represented the variation of τ as a function of the inverse of the maximum speed $V_M(T)$.

Extrapolation of the curves $\tau = f\left(\frac{1}{V_M}\right)$, shown in Figure 23 for various

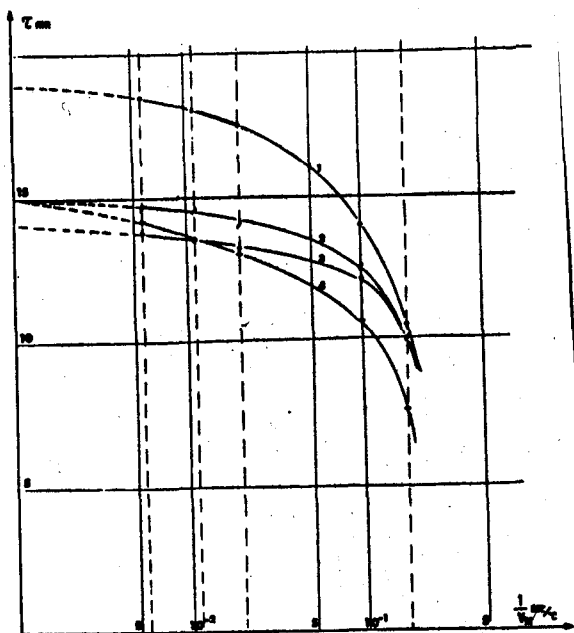


Figure 23. W 5% Re/W 26% Re.

- 1) Bare Wires, Diameter 0.5 mm.
- 2) Sheath: Ta, Diameter 1.3 mm.
- 3) Sheath: Ta, Diameter 1.8 mm.
- 4) Sheath: Ta, Diameter 1.8 mm.

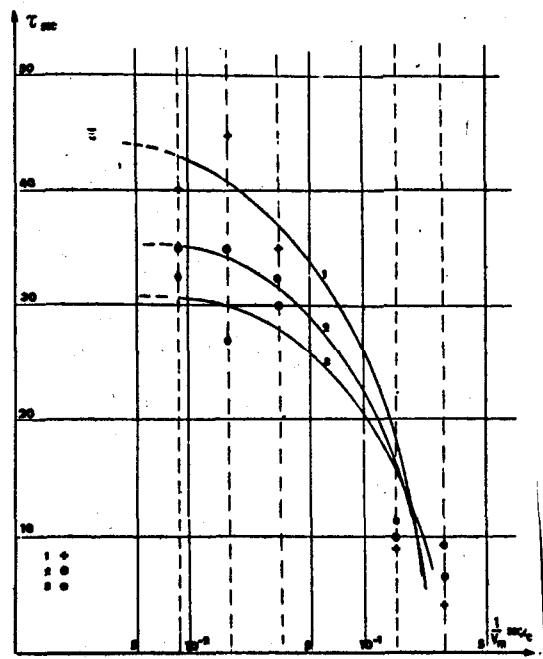


Figure 24. W 5% Re/W 26% Re.

- 1) Bare Wires, Diameter 0.5 mm.
- 2) Sheath: Ta, Diameter 1.3 mm.
- 3) Sheath: Ta, Diameter 1.8 mm.

types of thermocouples, gives the value of τ_0 to an accuracy on the order of one minute.

The values obtained under the conditions defined ($T_{\max} = 2000^\circ\text{C}$, $T_{\min} = 250^\circ\text{C}$) are as follows:

- τ_0 between 13 and 15 minutes for sheathed thermocouples (external diameter 1.8 or 1.3 mm) averaging 350 mm in length, and
- τ_0 between 18 and 20 minutes for bare wire thermocouples (diameter 0.5 mm) 250 mm long and insulated with refractory beads.

b) Time of Response to Heating. -- The presence at the end of the range of a zone in which the temperature gradient is not linear falsifies the very principle of measurement of the time of response to heating. Nevertheless, as an indication, we have shown in Figure 24 the values of τ as a function of the inverse of the mean speed $V_m(T)$. It may be supposed that if the conditions of linearity had been satisfied the response times would have increased considerably.

In Part One we described the principle of the cold welding compensation of a thermocouple.

There are alloys (X-Y) already on the market which permit compensating tungsten-rhenium thermocouples up to a junction temperature near 450°C. These alloys have the disadvantage, however, of exhibiting great resistivity, the effect of which is more marked the greater the lengths used.

That is why we have tried to develop alloys simple in composition and therefore easily reproducible, possessing low resistivity, and providing for the compensation of thermocouples of the W 5% Re/W 26% Re type, which are most used.

I. EXPERIMENTAL APPARATUS

We have seen that to determine the combination of two alloys such as P and Q (Figure 5), intended to provide compensation, it suffices to measure the emf delivered between T° and T_j by the thermocouple made up of these two elements.

As the junction temperatures T_j are rarely higher than 500°C, we put the alloys to be studied into a straight furnace with an atmosphere of argon.

II. STUDY OF THE ALLOYS

1. Preliminary Tests

After various not very conclusive tests done with materials currently used in thermoelectricity (chromel, alumel, iron, constantan, copper-nickel, etc.), bibliographic research led us to study binary cobalt-iron alloys. /54

Measurements done with numerous different combinations of these alloys allow the conclusion that the best compensation is obtained with the following percentages: Co 23.5% Fe and Co 17% Fe, the first in combination with W 5% Re and the second with W 26% Re.

Figure 25 permits comparison of the emf's delivered by thermocouples of Co 23.5% Fe/Co 17% Fe, W 5% Re/W 26% Re, and combinations of these.

2. Characteristics of the Thermocouple Co 23.5% Fe/Co 17% Fe

a) *Cobalt-Iron Equilibrium Diagram.* -- The equilibrium diagram shows that these alloys are situated in a solid phase whose first transformation line is at 700°C for the alloy containing 17% iron and at 850°C for that containing 23.5% iron.

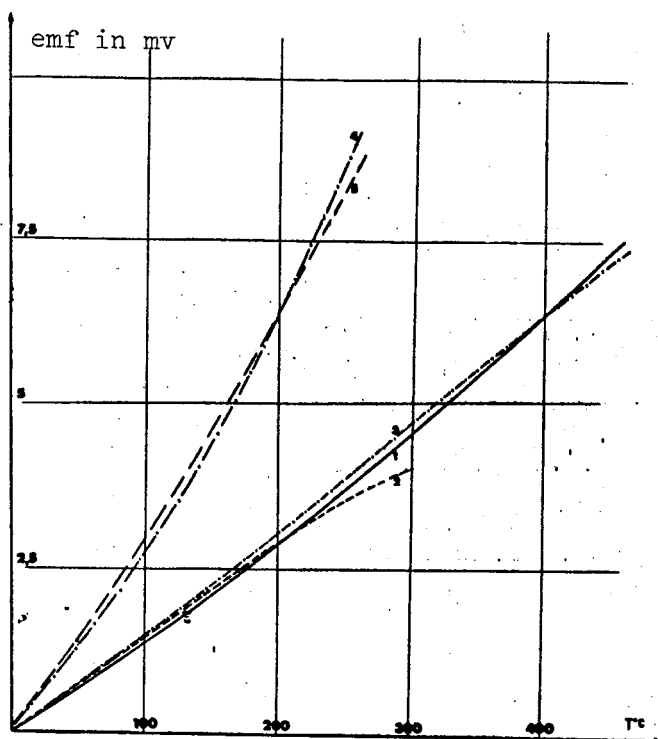


Figure 25. 1) W 5% Re/W 26% Re;
2) Co 23.5% Fe/Co 17% Fe; 3) X - Y;
4) W 26% Re/Co 17% Fe; 5) W 5% Re/Co
23.5% Fe.

a) *Resistivity of the Wires.* -- Measurements of resistivity at 20°C produced the following values:

Co 23.5% Fe: $10 \cdot 10^{-6}$ ohms per cm ($12.2 \cdot 10^{-6}$ ohm/cm for W 5% Re)
Co 27 % Fe: $14 \cdot 10^{-6}$ ohms per cm ($28.5 \cdot 10^{-6}$ ohm/cm for W 26% Re)

III. COMPARISON WITH X-Y WIRES

/57

1. *Precision and Extent of the Compensation*

We did experiments similar to those just described with the commercially marketed alloys X-Y, and the results obtained are shown in Figures 25, 26, and 28.

The emf delivered by the X-Y thermocouple is shown in Figure 25,

The difference $\Delta E = E_{T^0}^{T_j}(X-Y) - E_{T^0}^{T_j}(W-Re)$ is shown in Figure 26, and

Figure 28 shows the error occurring in measurement of the temperature T of the hot weld.

b) *Precision and Extent of the Compensation.* -- The variations in difference of emf $\Delta E = E_{T^0}^{T_j}(Co,Fe) - E_{T^0}^{T_j}(W,Re)$ existing between this thermocouple and the W 5% Re/W 26% Re thermocouple are illustrated in Figure 26.

Figure 27 represents the error occurring in the measurement of the temperature T of the hot weld as a function of T and for various temperatures of the junction T_j , the alloys being mounted as we have just described.

These curves permit us to state that this type of alloy affords an excellent compensation of the W 5% Re/W 26% Re thermocouple up to a junction temperature of about 250°C, since the maximum error is 1% for $T = 2000^\circ\text{C}$ and $T_j = 250^\circ\text{C}$.

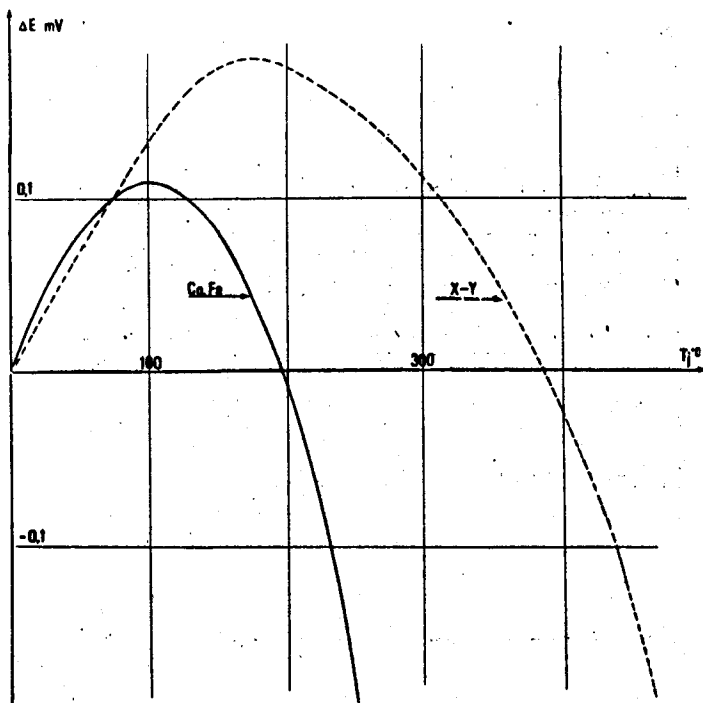


Figure 26.

2) *Resistivity of the Wires.* -- The resistivities at 20°C as given by the manufacturer are equal to:

$86 \cdot 10^{-6}$ ohm/cm for the wire connected to W 5% Re, and $115 \cdot 10^{-6}$ ohm/cm for the wire connected to W 26% Re.

IV. CONCLUSION

We find that to effect the cold welding compensation for a W 5% Re/W 26% Re thermocouple it is preferable to use cobalt-iron alloys when the junction temperatures do not exceed 250°C and the wire lengths used are great; on the other hand, the use of X-Y alloys is found to be more effective for junction temperatures which go up to 450°C and for wires of short length.

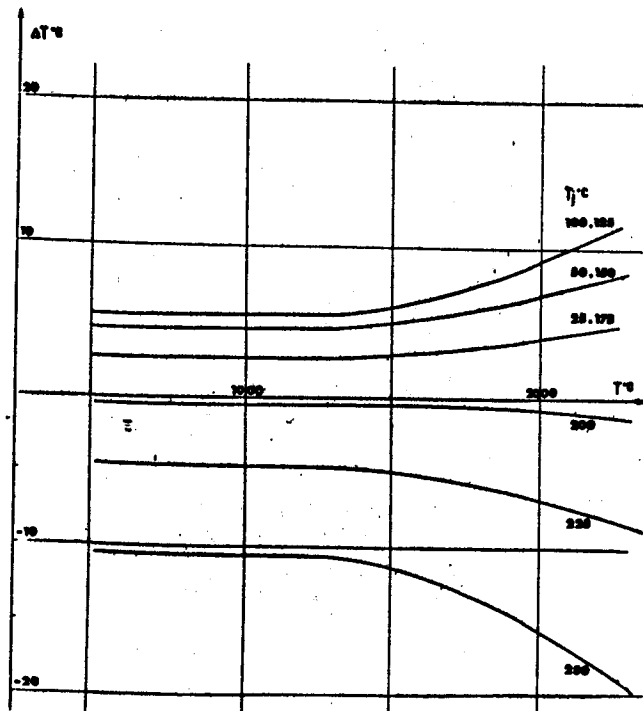


Figure 27. Fe-Co Alloy Wires.

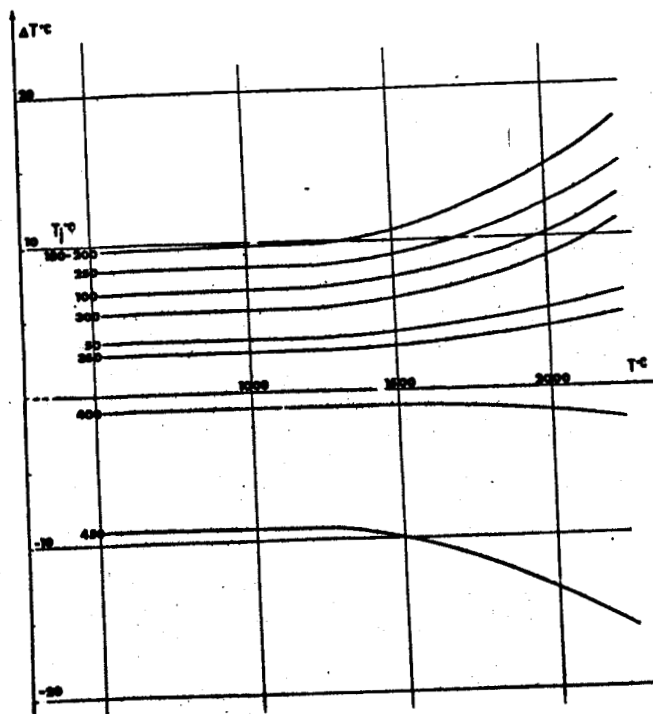


Figure 28. X-Y Wires.

STUDY OF TUNGSTEN-RHENIUM ALLOY THERMOCOUPLES
INSIDE THE REACTOR

The difficulty of taking account directly of the evolution of thermocouples in the course of irradiations in the reactor over an extended range of temperature led us to undertake a manipulation based in principle on a comparative study. The different phases of that manipulation are as follows: /61

The thermocouples contained in a sheath (G₂) are calibrated in a first furnace (TETARD),

They are then subjected, inside a second furnace (HEBE), to a series of irradiations at high temperature, and

Finally, the thermocouples are put back into the first furnace to be recalibrated.

It is then possible by comparing the characteristics found before and after irradiation to deduce the influence of such treatment on the emf delivered by the thermocouples.

A. EXPERIMENTAL ARRANGEMENT

This experimentation implies the setting up of a relatively extensive experimental apparatus, which must also meet very definite standards, for the handling of irradiated products necessitates certain precautions, and the operations in the reactor are subject to very strict rules.

This apparatus consists of:

The reactor SILOE, where the irradiations took place,

The irradiation furnace HEBE, belonging to the Grenoble center, which is installed inside the reactor,

The calibration furnace TETARD, located on a fixed base in one of the /64 operating wells of the reactor,

The G₂ sheaths containing thermocouples to be irradiated, and

The auxiliary equipment -- electric power supply, pumping unit, recording devices, etc.

I. THE REACTOR SILOE AND THE FURNACE HEBE

1) *The Reactor SILOE*

It will be recalled that this is a reactor of the "open-heart, swimming pool" type capable of attaining an operating power of 15 Mw, with the following fluxes:

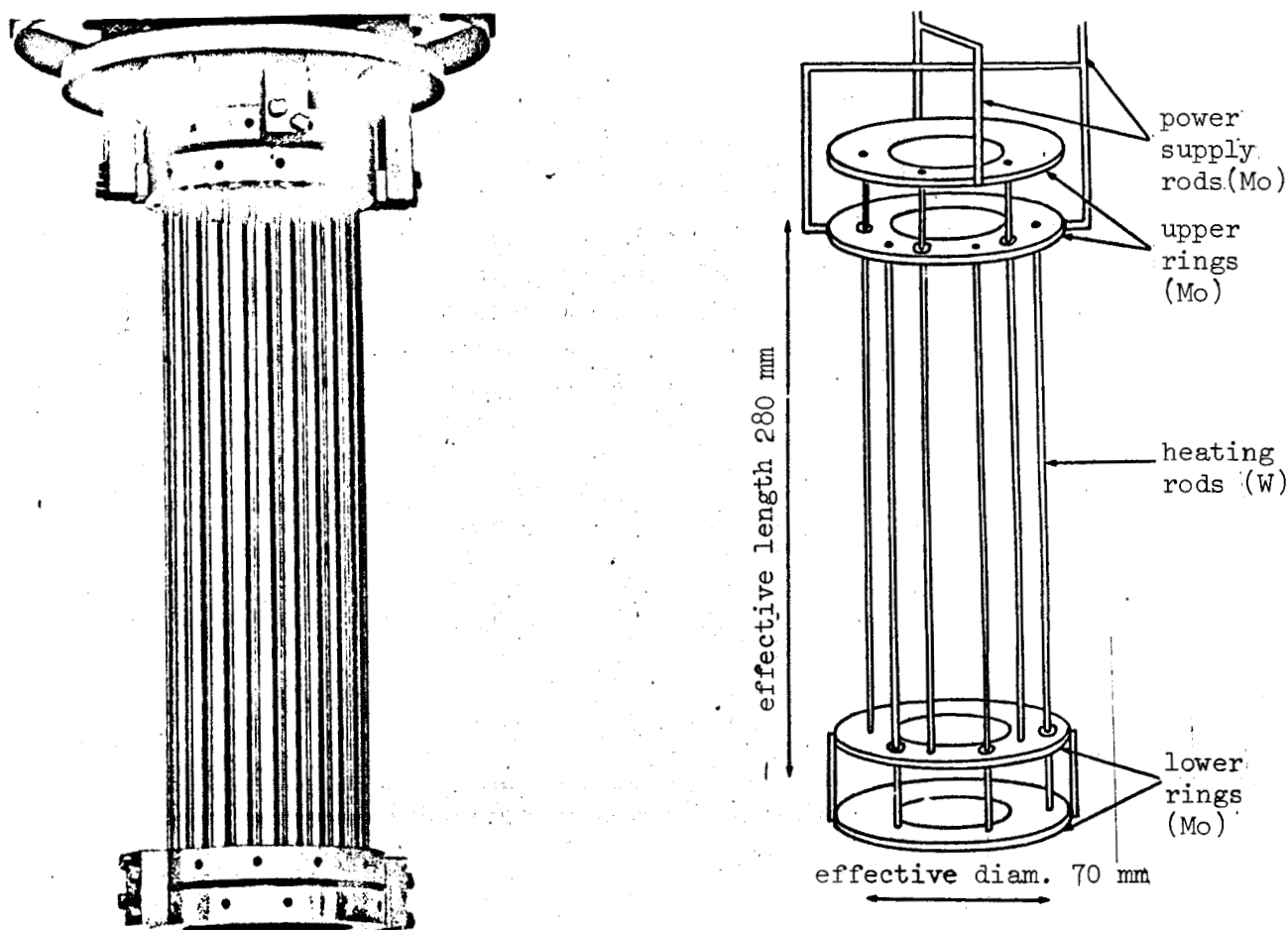


Figure 29. Heating Element of the Furnace TETARD.

Mean thermal flux: 10^{14} neutrons \cdot cm $^{-2}$ \cdot sec $^{-1}$

Mean rapid flux: $1.5 \cdot 10^{13}$ neutrons \cdot cm $^{-2}$ \cdot sec $^{-1}$

Its 25 fuel elements (uranium enriched to 90%) and 5 control elements with 10 uranium plates and a boron carbide bar ensure its continuous functioning for 3 weeks at normal operating power.

Thirty positions around the heart, for twenty-odd rings or capsules, are available to experimenters.

2) The Irradiation Furnace HEBE [31]

Developed and built at the Grenoble center, this furnace is designed for irradiation of non-fissile elements between 400°C and 1400°C under an inert atmosphere. Although a temperature of 1000°C represents a limit for its heating elements, nuclear heating enables the specimens, in this case the sheath G₂, to attain the temperature of 1300°C planned for irradiation of the thermocouples.

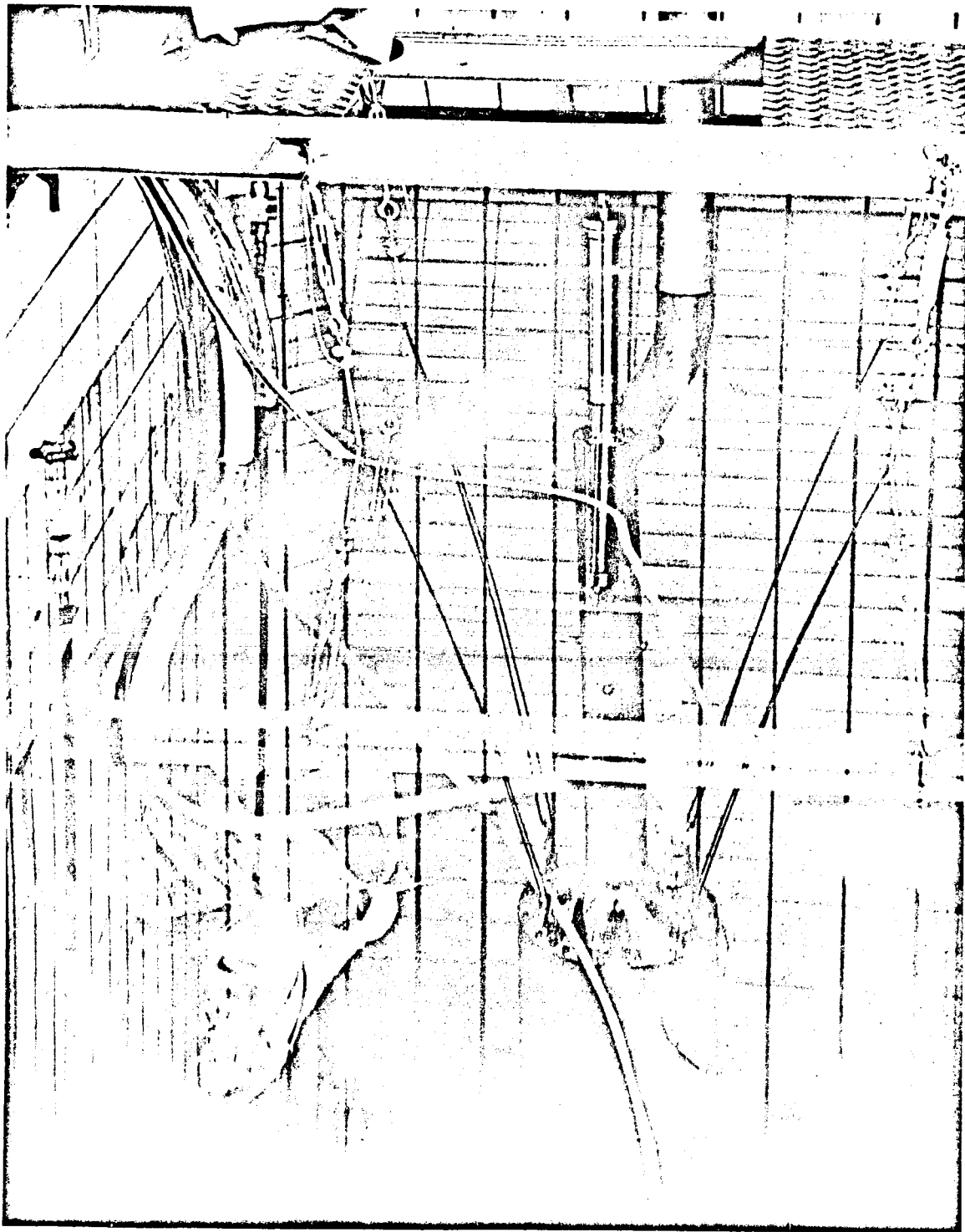


Figure 30. The Furnace TETARD in the Reactor.

II. THE CALIBRATION FURNACE TETARD

The design criteria of the furnace TETARD are such that it should:

Consume the power necessary to obtain a temperature of 2000°C by continuous variation, assuring good thermal stability for measurements,

Be vacuum-tight -- functioning at 10^{-5} mm Hg, in dynamic operation, being indispensable,

Be immersible in water,

Permit easy reading of the temperature of the specimens,

/65

Accomplish the installation of the sheath G₂ without risk,

Have a hydrodynamic form facilitating its cooling by natural convection of the water of the swimming pool,

Be of dimensions suited to rapid pumping and functional installation.

1. Description of the Furnace

a) *The Heating Element.* -- 24 tungsten rods (diameter 2 mm, length 275 mm) connected to molybdenum rings and arranged as shown in Figure 29 constitute the heating element.

Such an installation permits the resistor to expand without restriction, while at the same time avoiding field effects. In addition we may assume that the temperature along the axis is constant for some ten centimeters on each side of the center.

b) *The Current Feed-In.* -- Four molybdenum bars symmetrically distributed on the rings, fixed two by two to gas-tight openings, provide for the current feed-in. Two braided copper cables (diameter 25 mm) connected to the openings supply the electric power.

c) *The Thermal Screens.* -- To the number of six, these (4 of molybdenum and 2 of stainless steel) are arranged around the heating element in the same way as in the furnaces already described.

d) *The Outer Shell.* -- Designed as a single sheet of stainless steel (thickness 5 mm) and consequently without self-cooling, this shell is closed at the ends with cones which give it a hydrodynamic shape.

On the lower cone are two gas-tight outlets, one for the sheath containing the calibration thermocouples, the other for a vacuum gauge. /66

The dimensions of this unit (shell and cones) are as follows: height 830 mm, diameter 355 mm.

e) *The Pumping Sleeve.* -- This consists of a stainless steel cylinder (diameter 100 × 108, length 2 m) and ends in a chimney to which is fitted a funnel permitting the sheath G₂ to be introduced without impacts. A tube on the inside guides the sheath. Figure 30 shows the furnace immersed in the swimming pool.

2. Measurement of the Temperature

The temperature is measured by means of tungsten-rhenium alloys previously calibrated outside the reactor and placed in the G_1 sheath (container of calibration thermocouples) installed to stay in the furnace.

3. Installation of the Furnace

In order to protect the sheath G_2 from radioactivity when, once irradiated, it is introduced into the furnace TETARD, the latter is placed 3 meters deep in the water of the swimming pool.

The maintenance of the furnace is ensured by cables fixed to a catwalk installed over the swimming pool. This system permits easy access from the chimney, the opening of which is several centimeters above the level of the water. The various connecting wires are enclosed in an impervious sheath.

4. Characteristics of the Furnace

Figure 31 shows as a function of the temperature the variations of the power consumed and the intensity passing through the resistance.

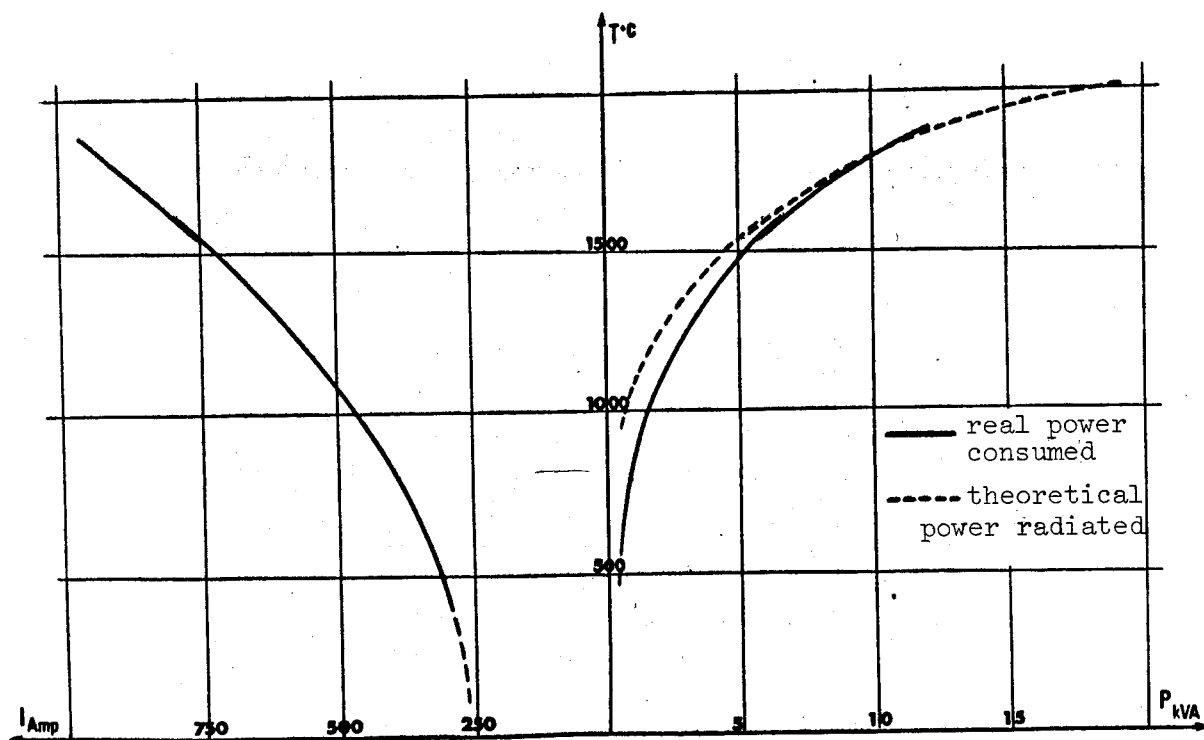


Figure 31.

The curve of Figure 32 shows the thermal inertia of the furnace by bringing out the variation of the temperature of the sheath G_2 as a function of the time, the heating element not being supplied with current past 1500°C.

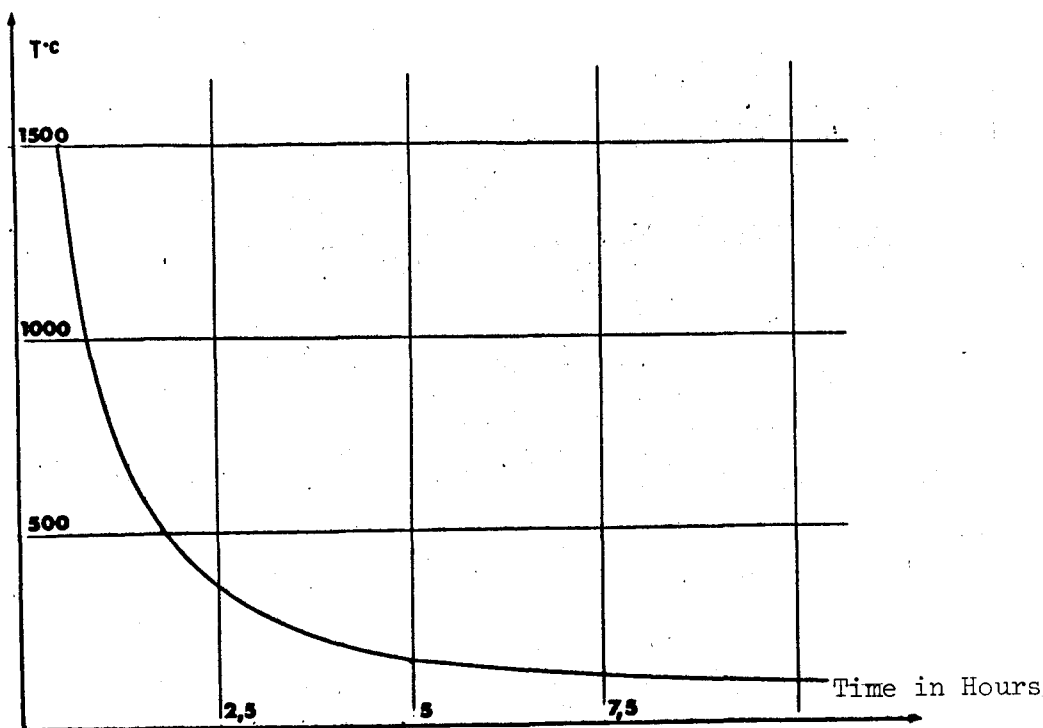


Figure 32. Cooling Curve.

III. THE SHEATHS CONTAINING THE THERMOCOUPLES

/69

In order for the calibration and specimen thermocouples to be under identical thermal conditions, the hot parts of the sheaths which contain them must have the same configuration. Their configuration has been worked out according to criteria based on the use of the sheath designed for the specimens, which, to be specific, should:

Have dimensions determined by the nuclear heating and the permissible size in the furnace HEBE,

Permit the installation of the maximum number of thermocouples,

Be made of materials providing good mechanical and thermal properties at 2000°C, and

Facilitate the various installation and maintenance work and adjustments.

1. The Sheath (G_2) Containing the Specimen Thermocouples

a) The Hot Zone. -- This consists of:

A central column supporting the axial stresses,

A body consisting of four cylindrical slots to receive the thermocouples,

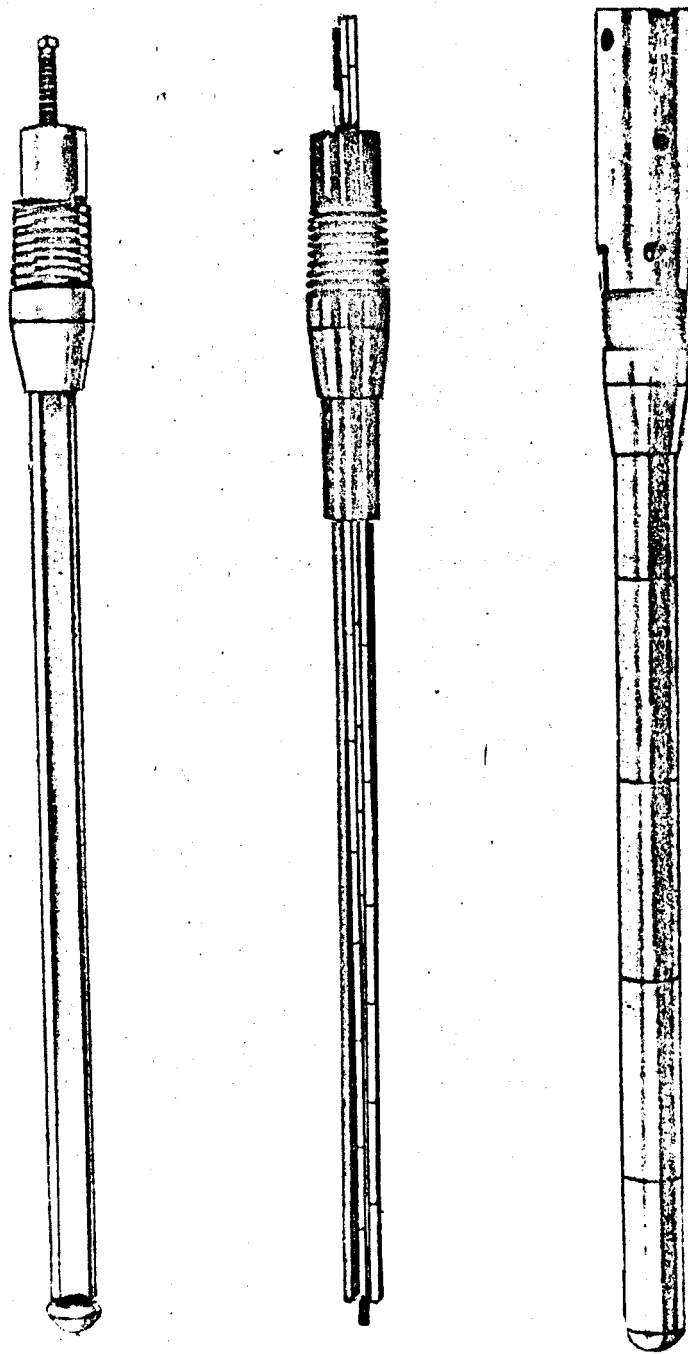
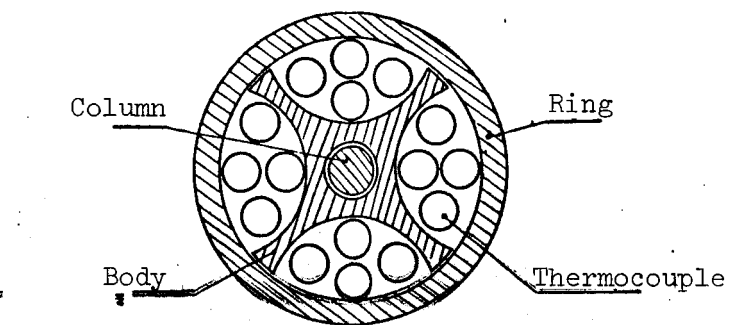


Figure 33. Sheath Containing the Thermocouples.

A series of rings surrounding the unit to provide rigidity, and
A hemispherical cap closing the lower end.

The choice of molybdenum as a material for these pieces and the presence of the thermocouples and of insulating beads give the whole a mass of 45 g/cm with the following dimensions:

Outside diameter: 30 mm

Length (determined by the rate of the neutron flux): 600 mm.

Figure 33 shows this part of the sheath in the course of various stages of assembly, together with a cross-section diagram. Such a design permits the installation of 16 thermocouples of a maximum diameter of 4 mm.

b) The Connecting Tube. -- The hot part is fixed to a stainless steel tube (diameter 36 × 40 mm, length 2,150 mm) by a bayonet device which eliminates projecting pieces which would impede the smooth introduction of the /70 sheath into the furnace and ensures an excellent rigidity and small bulk of the entire unit.

c) The Connecting Device. -- The wires of the thermocouples are soldered to the ends of a SOCAPEX block with 39 channels fixed to a box connected to the connecting tube, gas-tightness being effected by a coating of araldite.

2. The Sheath (G₁) Containing the Calibration Thermocouples

This consists entirely of a hot part (length 250 mm) and a connecting device identical to those of the sheath just described.

IV. THE AUXILIARY DEVICES

1. The Pumping Unit

Maintenance of a vacuum with a pressure less than 10^{-3} mm Hg in a volume of some hundred liters (furnace, sleeve, ductwork) during degasifications at high temperature requires the use of a relatively large pumping unit, consisting of a primary blade-type pump with a capacity of 60 m³/h and a secondary oil-diffusion pump of a capacity of 1000 l/sec.

The pressure is measured by means of two gauges, one mounted directly on the pumping unit and the other at the bottom end of the furnace.

2. Electric Power Supply

A saturable self-induction device outputs to a voltage-reducing transformer which feeds the heating element at 20 volts, 1000 amperes for the maximum power.

3. Measurement of the Electromotive Force

As in the preceding cases, an MECI recording instrument, Speedomax

type with 12 tracks, provides a record of the emf. A commutation system permits distribution of the various thermocouples over the 12 tracks available.

B. EXPERIMENTATION

/71

I. GENERAL

In order to impart a statistical character to the study of the behavior of thermocouples under flux, we carried out two series of irradiations solely on W 5% Re/W 26% Re thermocouples.

1. *Chronology*

In accordance with the principle of the undertaking, the various operations took place in the following order:

a) *First Series of Irradiations:*

Precalibration,

First irradiation cycle (3 weeks),

Transfer and calibrations (E_1 and E'_1),

Second irradiation cycle (3 weeks),

Shutdown of the reactor (1 week),

Third irradiation cycle (2 weeks),

Shutdown of the reactor and deactivation (4 weeks),

Transfer and calibration (E_2 and E'_2),

Dosimetry.

b) *Second Series of Irradiations:*

Precalibration,

First irradiation cycle (3 weeks),

Shutdown of the reactor (1 week),

Second irradiation cycle (3 weeks),

Shutdown of the reactor (1 week),

Third irradiation cycle (3 weeks),

Shutdown of the reactor and deactivation (20 weeks),

Transfer.

2. *Distribution of the Thermocouples*

With the cells provided in the G₂ sheath, the following thermocouples were irradiated:

/72

a) First Series of Irradiations:

- 4 of bare wires diameter 0.5 mm (N 0.5)
- 2 of bare wires diameter 0.3 mm (N 0.3)
- 5 tantalum sheathed, external diameter 1.3 mm (G 1.3)
- 1 tantalum sheathed, external diameter 1.8 mm (G 1.8)

b) Second Series of Irradiations:

- 2 of bare wires diameter 0.5 mm
- 1 of bare wires diameter 0.3 mm
- 2 tantalum sheathed, external diameter 1.8 mm
- 4 tantalum sheathed, external diameter 1.3 mm
- 2 niobium sheathed, external diameter 1.8 mm
- 2 sheathed with tantalum and Schoop-sprayed with molybdenum, external diameter 1.8 mm
- 1 of bare wires diameter 0.5 mm (molybdenum/niobium)

3. Remarks

a) We shall see later that the conditions under which the irradiations were carried out had the effect of bringing about, during the process, a release of oxygen in the furnace HEBE. This led to the formation of tantalum oxide in a powdery, extremely radioactive form. For that reason the activity of the sheath containing the specimen thermocouples and the contamination that it brought about compelled us to:

Reduce the number of transfers of the sheath between the furnaces HEBE and TETARD, i.e. the number of calibrations after irradiation,

Diminish the presence or the action of tantalum in the second series /73 of irradiations by Schoop-spraying the tantalum sheaths with molybdenum, or by the use of niobium sheaths,

Increase the time between the end of an irradiation and the calibration so as to lower the activity of the elements contained in the sheath G₂ by natural decay.

b) The measurement of the dosimetry is common to the two series of irradiations, since the respective locations of the furnace within the reactor have identical characteristics.

II. THE PRECALIBRATIONS

Before irradiation, the specimen thermocouples, previously subjected to annealing, were calibrated between 1000°C and 1850°C by means of the thermocouples contained in the sheath G₁. The difficulty of obtaining a stabilization at low temperature that would guarantee an accurate reading of the emf compelled us to make measurements only above 1000°C. For safety reasons we fixed the maximum working temperature at 1850°C.

We did not take account of the temperature of the cold solder junction (temperature of the junction between thermocouple wires and connecting wires). In fact, since this remained constant it is pointless to reduce

the accuracy of the results by bringing in supplementary corrections, the measurements being essentially comparative.

III. THE IRRADIATIONS

After the precalibration the sheath containing the specimen thermocouples was placed in the furnace HEBE in such a way that its lower end was at the level of the median plane of the heart of the reactor (maximum flux).

1. Atmosphere

Although the furnace HEBE was placed under an atmosphere of pure helium at a constant pressure of 0.750 kg/cm^2 throughout the irradiations, we observed a considerable oxidation of the elements of the sheath G_2 . This oxidation was no doubt occasioned by the partial decomposition of the non-stoichiometric glucina and the liberation, under the influence of irradiation at high temperature, of the oxygen adsorbed to the stainless steel of the furnace HEBE.

2. Dosimetry

/75

a) *Neutron Flux.* -- The characteristics of the location chosen for the various irradiations were determined on a mock-up which faithfully duplicated the sheath G_2 and was installed under the same conditions. Dosimeters along the length of the mock-up made it possible to measure the thermal flux and the rapid flux. Figure 34 gives the value of these fluxes as a function of the distance from the median plane.

b) Time of Irradiation and Integrated Fluxes

The duration of the irradiations and the corresponding integrated fluxes break down as follows for the two series:

	First Series of Irradiations		Second Series of Irradiations	
	Time (sec.)	Diam _i ($n \cdot \text{cm}^{-2}$)	Time (sec.)	Diam _i ($n \cdot \text{cm}^{-2}$)
First Cycle	$1.75 \cdot 10^6$	$5.3 \cdot 10^{19}$	$1.79 \cdot 10^6$	$5.4 \cdot 10^{10}$
Second Cycle	$1.66 \cdot 10^6$	$5.0 \cdot 10^{19}$	$1.79 \cdot 10^6$	$5.4 \cdot 10^{19}$
Third Cycle	$1.23 \cdot 10^6$	$3.7 \cdot 10^{19}$	$1.60 \cdot 10^6$	$4.8 \cdot 10^{19}$
Total	$4.64 \cdot 10^6$	$1.4 \cdot 10^{20}$	$5.18 \cdot 10^6$	$1.6 \cdot 10^{20}$

3) Temperature of Irradiation

We recorded simultaneously the emf's delivered by the specimen thermocouples and the chromel/alumel thermocouples permanently installed in the furnace HEBE.

The mean temperatures measured by the specimen thermocouples were

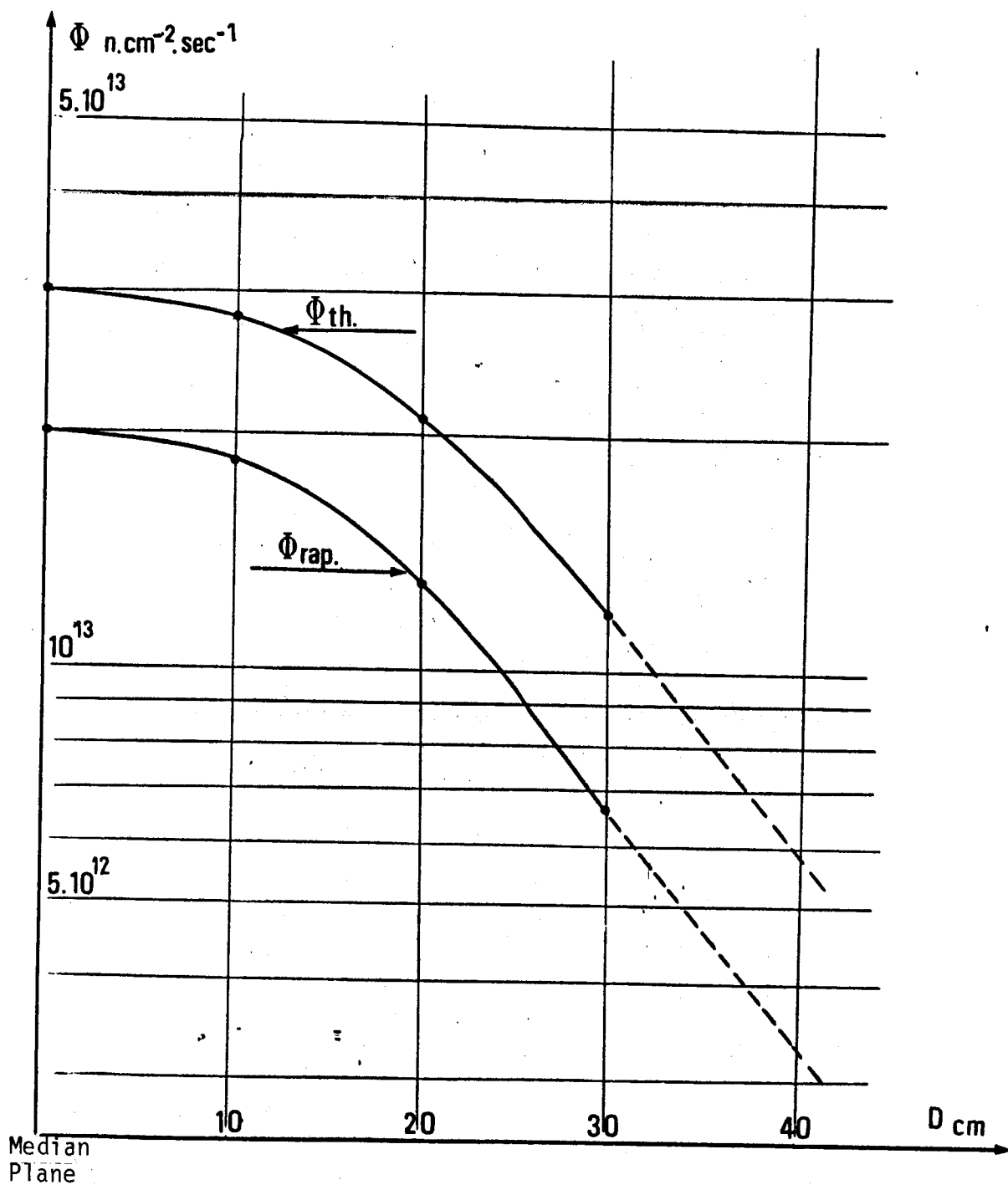


Figure 34. Flux Gradient.

about 1250°C for the first and second series of irradiations. The fluctuations in temperature observed in the course of a cycle on the recordings are lower than 1.5%.

The geometric configuration and the content of the sheath as well as the experimental conditions made it impossible to determine the thermal gradient of the thermocouples. However, as the heating was produced by the rapid flux, we may suppose that the rapid flux and the temperature were distributed similarly along the sheath.

IV. CALIBRATIONS AFTER IRRADIATION

/76

We carried out two calibrations (E_n and E'_n) of the irradiated thermocouples under the same conditions as the precalibration. A first heating after irradiation should in fact ensure the regeneration of the elements constituting the thermocouples and so eliminate the effect of phenomena of a reversible nature.

The measurements capable of giving an account of the behavior of irradiated thermocouples are the recordings during the irradiations and the curves of deviation. However, before going into their development it is necessary to point out the difficulties encountered during the various manipulations, which had the result of reducing considerably the number of results expected.

I. PRELIMINARY REMARKS

1. First Series of Irradiations

Defects in the manufacture of the thermocouples and the assembling of the sheath G₂ were reflected in anomalies or deviations in the curves of calibration made in the course of the first heating in the furnace TETARD. We were thus constrained to consider as the only valid indications those furnished by four (2 TC N 0.5, 1 TC G 1.3, and 1 TC N 0.3) of the twelve thermocouples installed, as only those four were in keeping with those of the calibration thermocouples.

2. Second Series of Irradiations

The operations of transferring the sheath G₂, subject to safety rules, were rendered considerably more complicated and difficult by the high radioactivity exhibited by that sheath even after the long period of deactivation (20 weeks). Moreover, the rapidity of the handling in transferring the sheath G₂ from the furnace HEBE to the furnace TETARD, weakened as it was by heating to high temperature (precalibrations) and the conditions of irradiation (oxidation), brought about a break at the level of the hot part of the molybdenum, thus suppressing any possibility of calibration after irradiation.

II. RECORDINGS DURING THE CALIBRATIONS

The temperatures indicated by the different specimen thermocouples being essentially identical (deviation <10°C attributable to the geometry of the sheath flux), we have entered in the table to Figure 35a the means and variations with respect to the means for temperatures corresponding to the emf's recorded in the course of the various cycles of the first and second series. A very rough comparison point is furnished by one of the regulative thermocouples (chromel/alumel) of the furnace HEBE, which by reason of its location best reflects the value of the flux.

The recorded curves for $\text{emf} = f(\text{time})$ are not shown; the reduction in scale did not make it feasible to show the behavior of the thermocouples properly. However, analysis of those curves and analysis of the table permit the formulation of the following statements:

		Specimen Thermocouples		HEBE Regulative Thermocouples	
		Mean Temperature in °C	Variations in Temperature ±°C	Mean Temperature in °C	Variations in Temperature ±°C
1st Series	1st Cycle	1270	10	815	1
	2nd Cycle	1250	20	810	7
	3rd Cycle	1250	13	805	15
2nd Series	1st Cycle	1265	10	815	7
	2nd Cycle	1230	10	830	7
	3rd Cycle	1150	15	810	2

Figure 35.

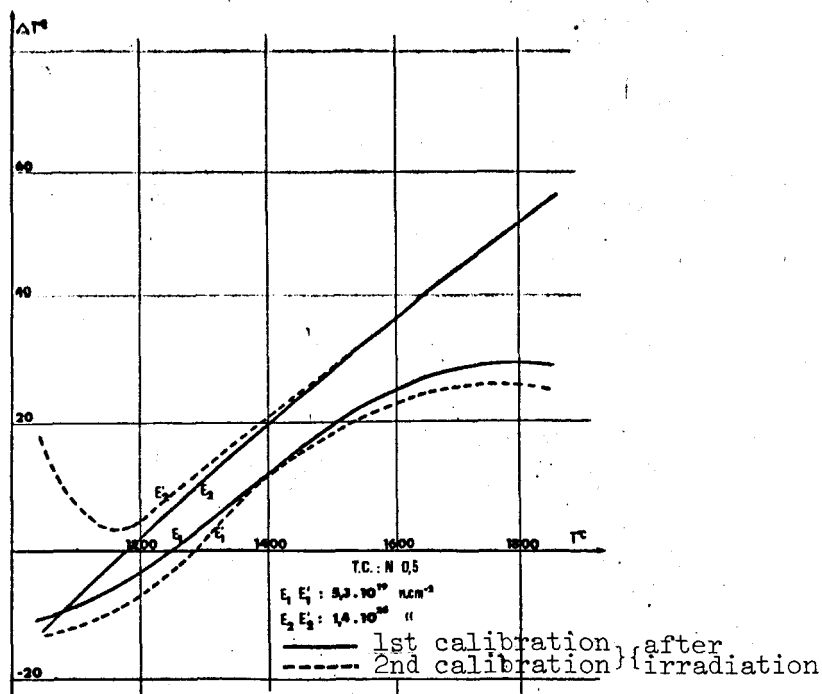


Figure 35 a.

- Temperature fluctuations followed in an identical manner by all the thermocouples and due to variations in flux mask any variation of the specimen thermocouples in the course of one and the same cycle,

- The temperatures attained at the end of each cycle are always very close ($\sim 3^\circ\text{C}$) to the mean temperatures observed,

The differences of the mean temperatures between two successive cycles should, first on the basis of the two notes offered above and second as a function of the values indicated by the HEBE thermocouple, justify the variation of the thermocouples in the course of time. In fact in the first irradiation series it shows no significant variation, in contrast to the curves of deviation (cf. § III). In the second series these differences in temperature are more marked, but it was not possible for us to compare them to those furnished by the corresponding deviation curves, since no calibrations were done after irradiation.

III. THE CURVES OF DEVIATION

It will be remembered that (page 23) the deviation curves represent for a given thermocouple the difference $\Delta T^\circ = f(T^\circ C)$ between the precalibration and the various calibrations after irradiation.

Figures 35a, 35b, and 35c show the deviation curves obtained for the four thermocouples of the first series (the two thermocouples consisting of bare wires 0.5 mm in diameter providing identical values).

We find from these results that:

The evolution of the thermocouples manifests itself in all cases by /81 a diminution of the emf delivered above 1300°C (this value corresponding approximately to the temperature of irradiation),

The deviation increases with the time of irradiation and is larger the smaller the diameter of the wires of the thermocouple, and

The influence of the first heating or calibration after irradiation (intended to ensure regeneration) is less significant as the duration of the irradiation increases.

IV. CONCLUSION

The absence of deviation curves in the second series of irradiations deprived us of a certain number of data that would no doubt have allowed us to give a statistical character to this study. Nevertheless, the reproducibility of the measurements collected and the observations made on the basis of these leads us to think that the reversible and irreversible phenomena which modified the thermocouples are multiple and that their relative influences cannot always be readily differentiated.

Before studying the various factors that contribute to the evolution of thermocouples irradiated under the conditions described above, it is proper to review the part played by the relative locations and extents of the temperature gradient in which the thermocouple to be calibrated is placed and the zone where the modification of structure took place in the wires of which it is constituted. In fact the modifications that the thermocouple undergoes may or may not be traced depending on the limiting cases illustrated in Figure 36.

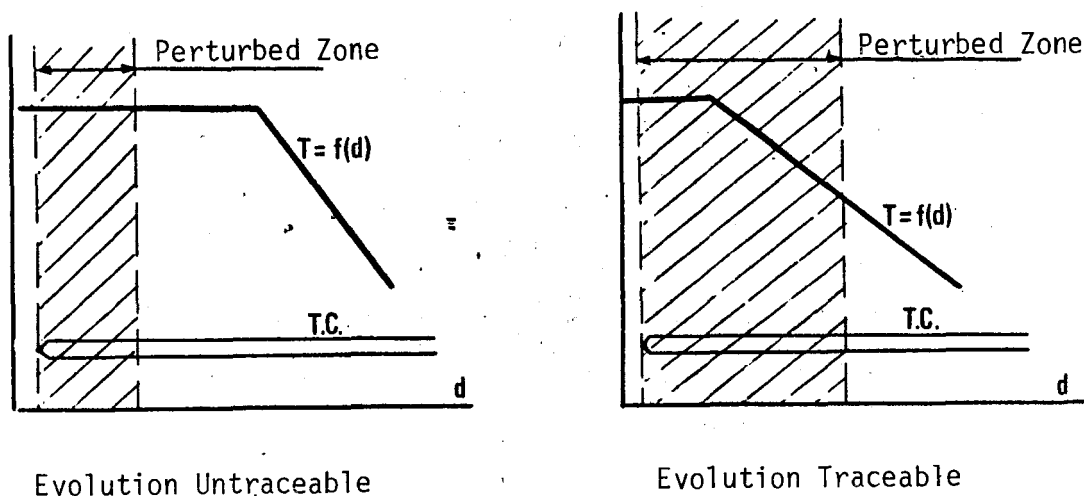


Figure 36.

I. TRANSMUTATIONS

Various authors [24 to 28] have shown that nuclear transformations, by reason of the modifications in composition that they induce within the irradiated materials, are one of the principal factors causing the evolution of thermocouples.

Theoretical studies have also been undertaken [32] to determine by Mott's [7] and Domenicalli's [8] formulas the deviations produced by these changes in composition in the emf's delivered by the thermocouples.

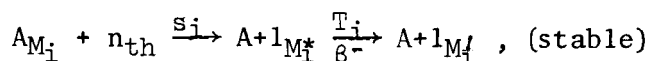
However, the difficulty of determining the energy distribution (flux = $f(\text{energy})$) and the geometrical distribution of the flux with respect to the specimen renders the exact calculation of the percentage of elements transmuted practically impossible. /84

In our study we have limited ourselves to the action of a unidirection-

al thermal flux of known energy (0.025 ev), in order to assign an order of magnitude to the compositions obtained after irradiation.

The principle of the calculation is as follows:

In a nuclear reaction of the type:



where A_{M_i} is one of the isotopes of the irradiated element A_M , the numbers of parts per million of $A+1_{M_i^*}$ and $A+1_{M_i'}$ formed during the period of irradiation t are given by:

$$\text{Number of ppm of } A+1_{M_i^*} = N = \frac{10^6 \cdot s_i \cdot n_i \cdot \phi_0}{s \cdot n^2 \cdot a \cdot L_i} \cdot [1 - \exp(-L_i \cdot t)] \cdot [1 - \exp(-s \cdot n \cdot a)]$$

$$\text{Number of ppm of } A+1_{M_i'} = N' = N \cdot \left[\frac{L_i \cdot t}{1 - \exp(-L_i \cdot t)} - 1 \right]$$

with n_i = number of atoms of A_{M_i} per cm^3

n = number of atoms of A_M per cm^3 ($n = \sum_i n_i$)

s_i = effective section of A_{M_i} of the thermal neutrons, in barns

s = mean effective section of A_M of the thermal neutrons, in barns

L_i = radioactive constant of $A+1_{M_i^*}$ ($L_i = 1/1.44 T_i$)

a = depth of the specimen of A_M traversed by the flux

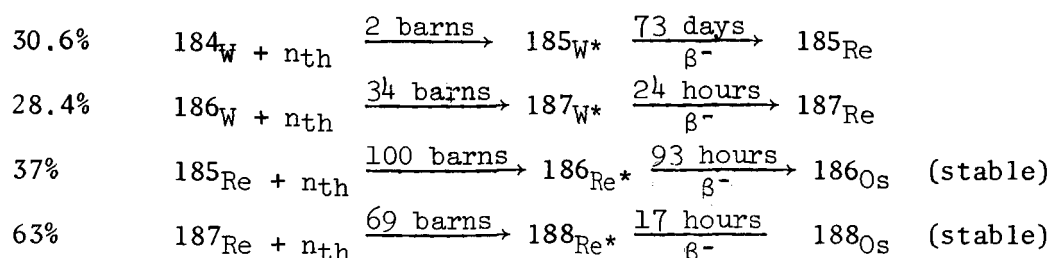
ϕ_0 = incident thermal flux in $\text{n} \cdot \text{cm}^{-2} \cdot \text{sec}^{-1}$

Note: In this calculation we have not taken account of the transmutations (regarded as negligible) of elements such as $A+1_{M_i'}$ and of the fact that $n_i = n_{i0} - m_i$, where n_{i0} is the number of atoms M_i of A_{M_i} before irradiation and m_i the number of atoms of $A+1_{M_i^*}$ formed; in fact, our experimental conditions permit us to make the approximation $n_i = n_{i0}$.

1. Nuclear Reactions

/86

Tungsten and rhenium being the sole elements constituting the wires of the thermocouples, the nuclear reactions capable of inducing significant changes in composition are the following:



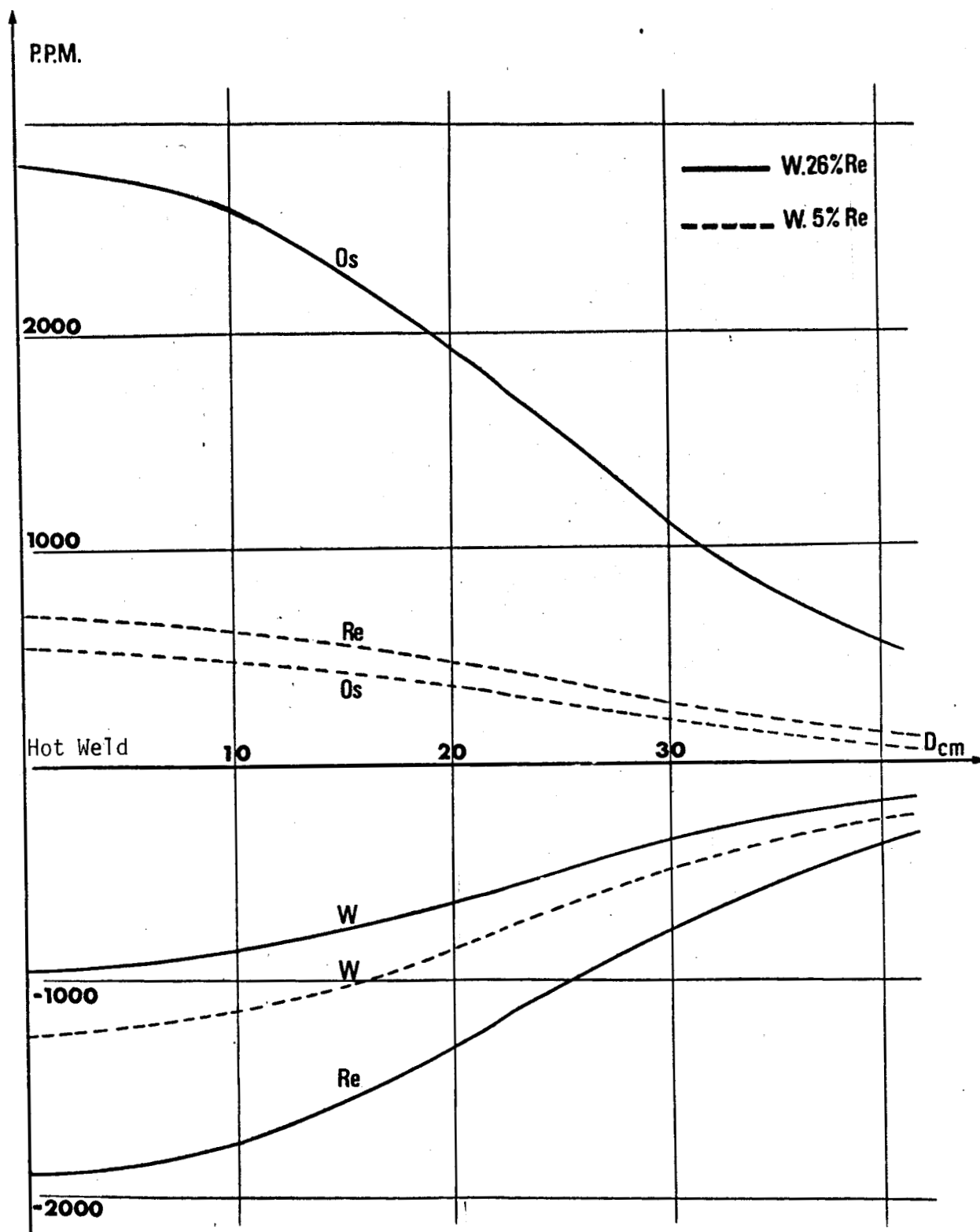


Figure 37. Variations in Composition.

2) Composition After Irradiation

Fig. 37 shows as a function of the distance from the median plane (level of the hot weld of the thermocouples) the variations in composition (in ppm) of wires of W 5% Re and W 26% Re (diameter 0.3 mm) with respect to the initial state, for 1300 hours of irradiation (first series) with the fluxes indicated above. These results are in perfect agreement with the curves $\Delta\% = f(\text{time, flux})$ calculated by D.N. Hall [33] for the same materials.

Notes:

The calculation for wires of 0.5 mm and 0.2 mm leads to largely identical values (deviation <3%).

It may be supposed that if we took account of the intermediate neutrons (whose distribution is unknown), which exhibit extremely high resonance sections for given energies and for that reason are rapidly attenuated, the variations in composition obtained for the various diameters of wires would lead to greater deviations.

A study done by using an isotropic flux, which is no doubt more suitable for the experimental conditions, would yield larger values.

3) Contribution to the Evolution of the Thermocouples

The irreversible character of the effects produced by transmutations and the fact that they are an increasing function of the time of irradiation correspond to the observations formulated concerning the deviation curves, especially as these effects are perfectly traceable throughout their extent.

II. PERTURBATION OF THE LATTICE

/ 87

1) Mechanism [34]

Irradiation at high temperature, by the creation of defects (vacancies, interstitial atoms, dislocations), induces perturbation of the lattice and so intensifies the process of diffusion of atoms. Nevertheless, the thermodynamic equilibrium toward which this perturbed state tends ensures the recombination of the vacancies and interstitial atoms as they are formed, but entails a change in the composition of the alloys by homogenizing them in the vicinity of the hot weld.

2) Effect on the Thermocouples

a) *Creation of Defects.* -- This operates throughout the zone in which the flux is applied. Nevertheless, the annihilation of the defects in the course of the irradiations and the annealing of the first calibration after irradiation should cause their effect (modification of the resistivity) to disappear completely, so that it will no longer be observable during the second calibration.

b) *Diffusion.* -- An essentially irreversible phenomenon, this affects only the immediate vicinity of the hot weld. In fact, the conditions of the irradiations (temperature and time) and the low values of the diffusion coefficients of tungsten and of the rhenium in the tungsten do not permit the creation of a zone of diffusion higher than the region where the tempera-

ture is constant in the heating element of the furnace TETARD (about 10 cm). For that reason, while it does contribute to changing the structure of the thermocouples, the effect of diffusion cannot be traced.

III. OTHER FACTORS

1) *Nuclear Heating*

As the calibrations after irradiation were done before the total deactivation of the elements of the sheath, the nuclear heating that they cause creates a thermal dissymetry between sheaths G_1 and G_2 . This phenomenon, which is significant at low temperature ($T < 250^\circ\text{C}$), has no effect at higher temperatures, where the power radiated by the resistor is far higher than that supplied in this way (~ 0.01 w).

2) *Oxidation*

/88

The presence of oxygen in the furnace HEBE during irradiations is capable of affecting the structure of the thermocouples by the formation of a surface coating of oxide on the wires. However, the irradiation temperature, distinctly higher than that of volatilization of the oxides, should ensure their disappearance as they are formed.

But this argument, which is valid for the hot parts of the sheath, is not valid in the colder zones, where the oxides cannot volatilize and consequently do cause a diffusion of oxygen in the lattice.

3) *Behavior of the Glucina*

Various works [35 to 37] have shown that glucina under irradiation is the subject of transformations (variation of the lattice parameters, diminution of the thermal conductivity, modification of the mechanical properties, etc.), which, however, are annihilated by heating outside the flux.

Such transformations, principally the drop in resistivity, may be much more significant in the case of sheathed thermocouples, where the glucina is in intimate contact with the wires, especially as the extent of the zone in which they are produced is greater than that of the region in which they can be eliminated.

BEHAVIOR OF THERMOCOUPLES OF REFRACTORY ALLOYS
UNDER IRRADIATION IN THE PRESENCE OF URANIUM DIOXIDE

The use of sheathed thermocouples at high temperature in UO_2 fuel elements under irradiation has been the subject of various works [38 to 40], of which it appears to us useful to give a brief résumé. In fact the use of these thermocouples under such conditions is limited by the influence of factors other than irradiation, the causes of which are not yet entirely clear.

1) *Observations Made*

It is found that the emf delivered by thermocouples before measuring the temperature of the fuel element in which they are placed decreases considerably after each rise in power of the reactor. This reproducible phenomenon does not at all modify their sensitivity. However, after a time of irradiation which varies widely depending on the experiment under consideration certain of them do not deliver any emf at all. Such a behavior, usually found between 1400°C and 2200°C , may be represented by the curves shown in Figure 38.

2) *Interpretation*

It is evident from these observations that in the presence of UO_2 two distinct phenomena appear:

a) *Deviation of the Electromotive Force.* -- As the deviations are far higher than those which can be induced by thermal shocks (starting up and dropping bars) and by irradiations, thermocouples can apparently be absolved, especially as their sensitivity is not modified and the deviations are reproducible. It appears that this phenomenon is chargeable to various factors which simultaneously cause a lowering of the temperature of the fuel element by loss of reactivity due to the poisoning of the UO_2 by xenon ^{/90} and samarium, and also a modification of the location of the thermocouple in consequence of expansions produced by the rise in temperature on starting up; for this reason the disturbance of the contact between thermocouple and UO_2 shows itself in a lower recorded temperature.

b) *Deterioration of the Thermocouple*

After irradiation the recovery of the fuel element permits the observation that thermocouples taken out of the circuit exhibit a corrosion of the sheath by UO_2 which causes their partial or total deterioration. This phenomenon is essentially a function of the temperature and time of irradiation and of the material constituting the sheath (molybdenum or tantalum). Of the two metals molybdenum appears to be less reactive.

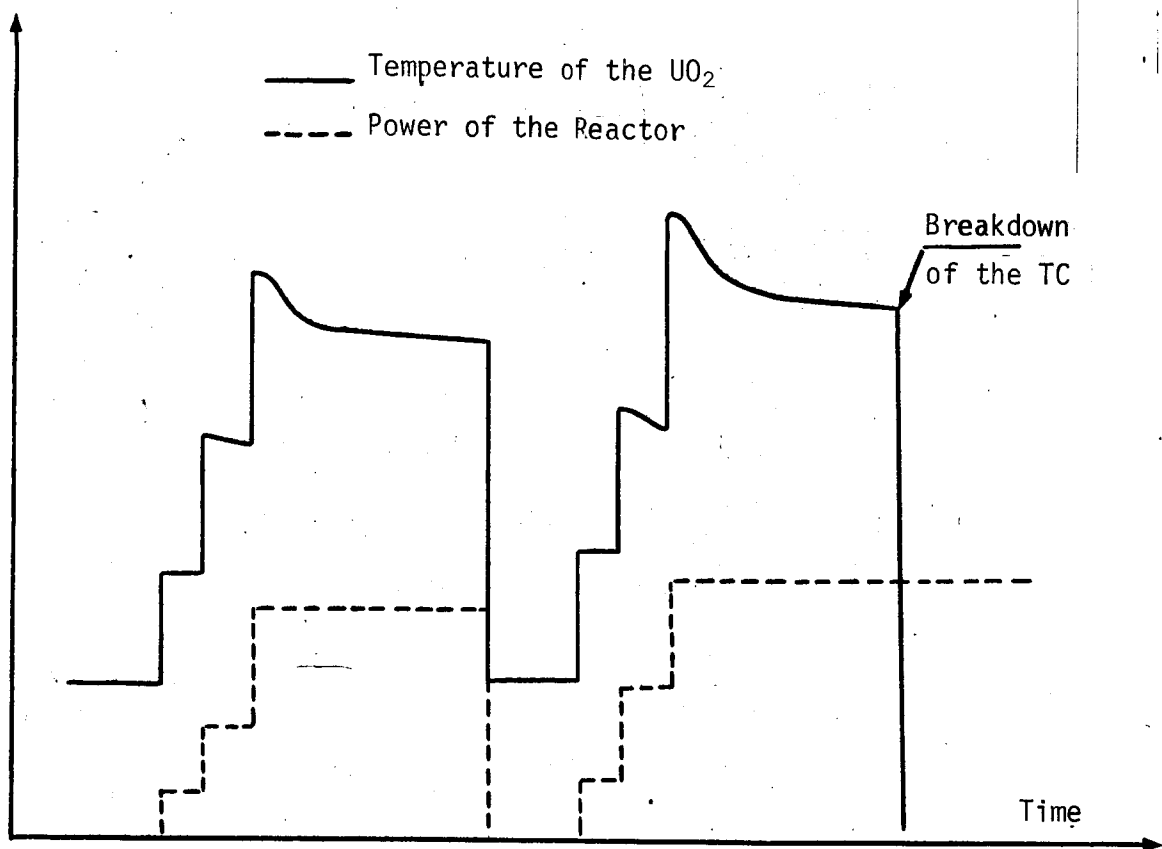


Figure 38.

Study of tungsten-rhenium alloy thermocouples outside the reactor proved to us that when used with certain precautions they constituted a particularly efficacious and precise means of measuring temperatures up to about 2300°C. In fact the emf's that they deliver, which are not altered in any way by long periods of heating, are very significantly modified by thermal shocks and by the presence of certain gases, but without that deviation's exceeding the limits of the zone of dispersion of purely thermoelectric origin. Moreover, the measurement of response time and the development of alloys which provide cold weld compensation make it possible to widen the field of numerous industrial applications of this type of thermocouple.

The results of study inside the reactor showed that W 5% Re/W 26% Re thermocouples under irradiation undergo considerable variations attributed principally to changes in composition caused by nuclear transmutations which limit the precision and the useful life of this type of thermocouple under neutronic fluxes. For that reason, from the point of view of prolonged use in the reactor it seemed to us useful to suggest a new type of thermocouple consisting of materials of very small cross section which exhibit a good compatibility with uranium dioxide and meet the criteria ordinarily required.

Determination of these substances *a priori* was provided for us by a comparison of the mean effective sections for thermal neutrons (in barns) of the refractory metals, listed in the table below. It will be noted that molybdenum and niobium show the lowest values.

Rhenium	Tantalum	Tungsten	Molybdenum	Niobium
84	21.3	19.2	2.5	1.1

In addition, study of the curves showing the variations of absolute thermoelectric power [41-42] and of resistivity [43] as a function of the 92 temperature, presented in Figures 39 and 40 respectively, permits us to state that molybdenum plays a rôle with respect to niobium identical to that of tungsten with respect to rhenium. Lastly, it should be noted that

1) Molybdenum and niobium, like tungsten and rhenium, occupy adjacent places in Mendeleev's periodic classification.

2) The molybdenum-niobium equilibrium diagram [44], given here as Figure 41, permits us to foresee a normal behavior of these thermocouples with a high percentage of molybdenum up to temperatures of the order of 2200°C, for it shows a continuous solid phase. (The solidus line reaches a minimum of 2345°C for a concentration of molybdenum of about 20% by weight.)

Thus we may suppose that molybdenum-niobium alloy thermocouples are capable of supplying emf's comparable to those of tungsten-rhenium alloys,

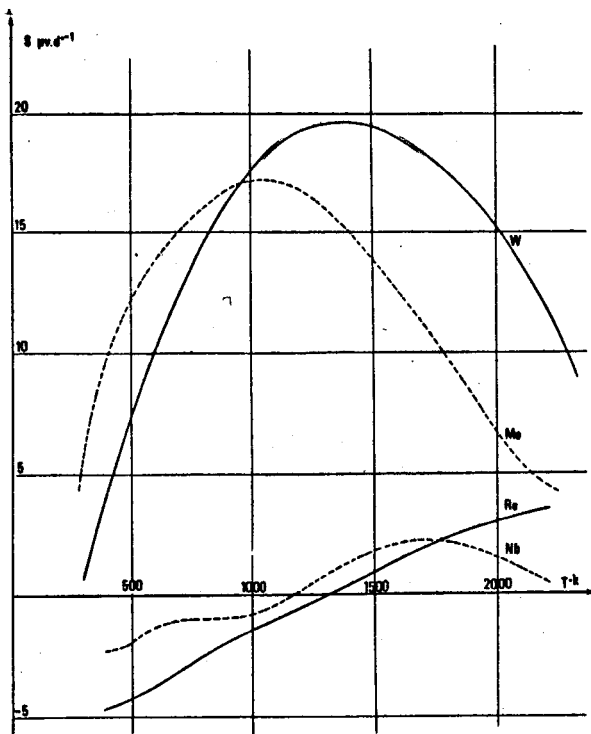


Figure 39. Absolute Thermoelectric Powers.

especially as the curves $E_{mv} = f(T^{\circ}C)$ (Figure 6) of thermocouples consisting of couples of the respective pure metals are largely identical.

The manufacture of such alloys poses metallurgical problems more complex than those of tungsten and rhenium [45]. It is possible, however, to make wires of various compositions in order to do preliminary calibrations for the purpose of determining the optimal conditions (highest and most nearly linear emf). It is useful to note that in the eventuality that pure molybdenum should realize that condition with some molybdenum-niobium alloy, thermocouples in the form of a coaxial sheath (of the type of Figure 7c) with a molybdenum-niobium core and the sheath of molybdenum would be perfectly suited to measurement of the temperature of UO_2 fuel elements.

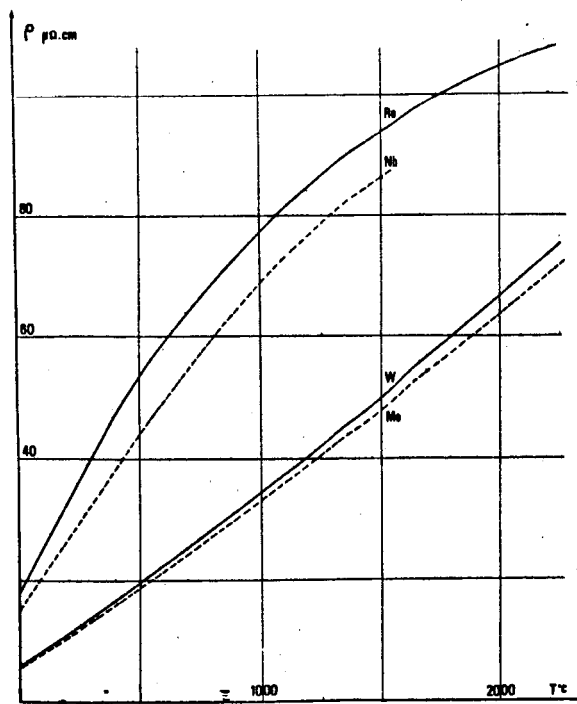


Figure 40. Resistivities.

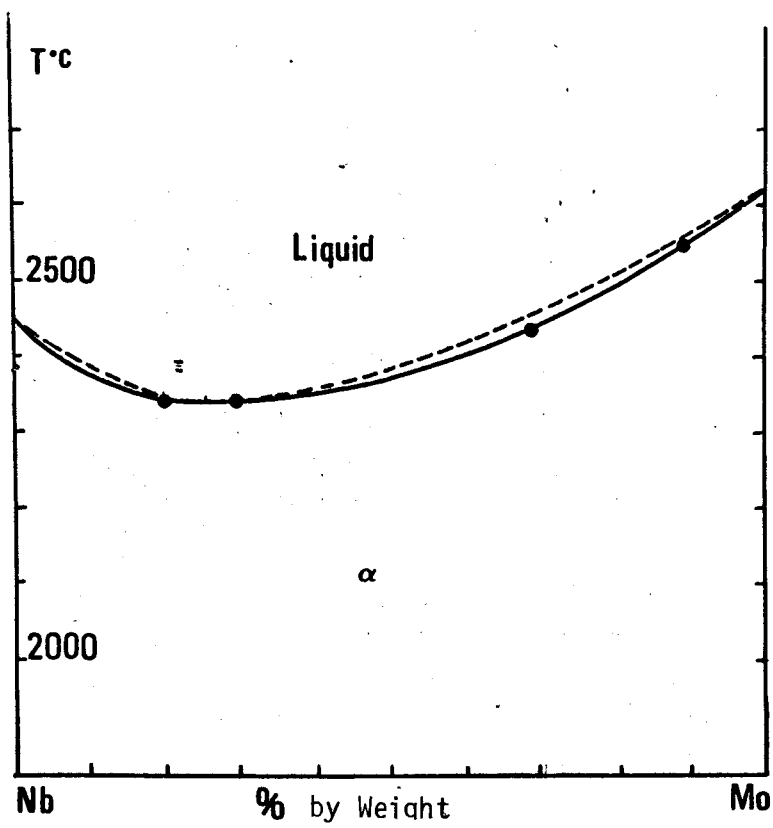


Figure 41. Mo-Nb Equilibrium Diagram.

TABLES AND MEAN CALIBRATION CURVES OF TUNGSTEN-RHENIUM ALLOY THERMOCOUPLES/95

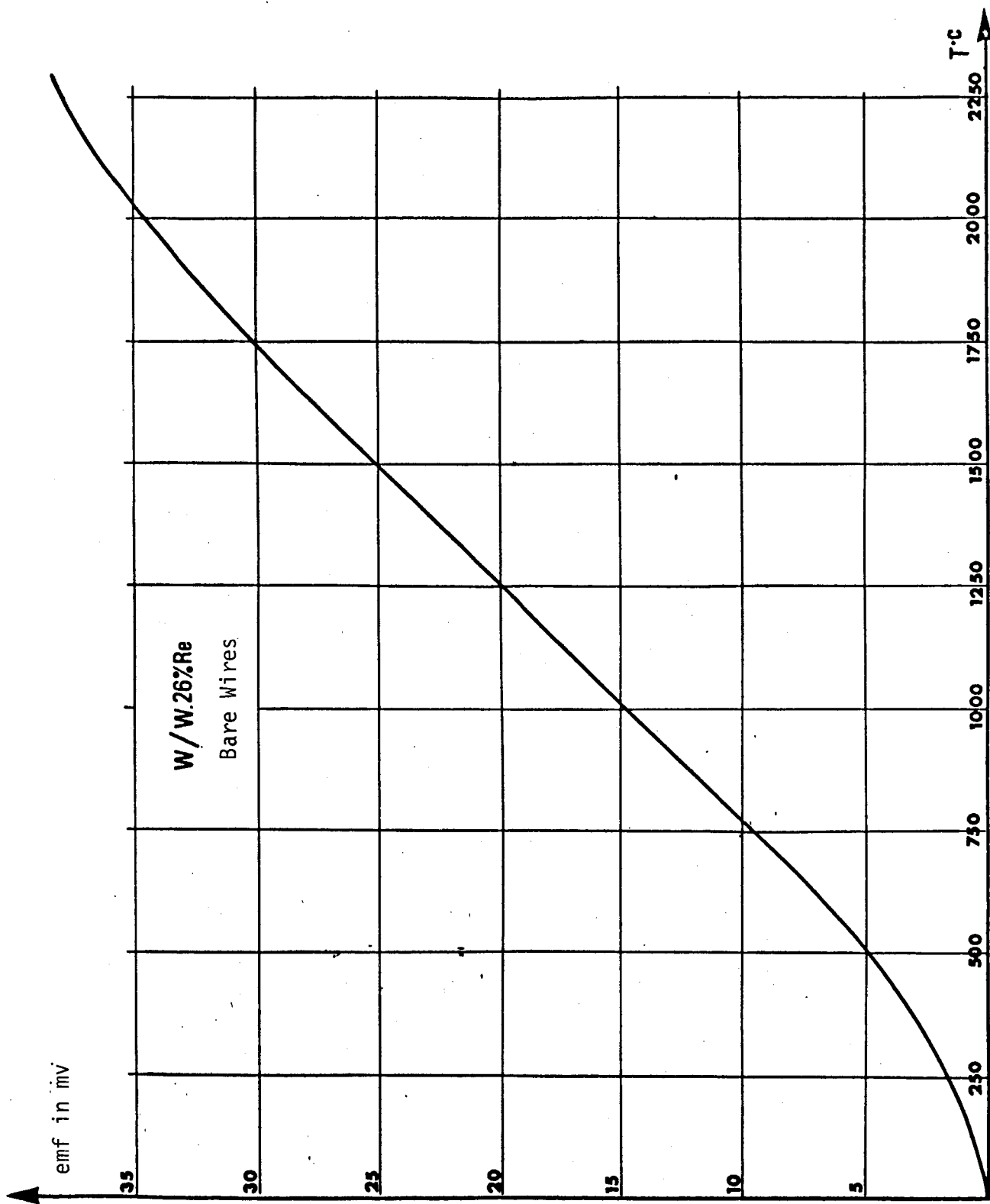
W/W 25% Re Bare Wires

EMF in mv :

Reference: 0°C

T°C	0	100	200	300	400	500	600	700	800	900	1000	1100	T°C
0	0,000	0,410	1,120	2,100	3,365	4,820	6,610	8,600	10,670	12,710	14,740	16,790	0
5	0,015	0,440	1,160	2,160	3,440	4,905	6,700	8,710	10,775	12,810	14,840	16,900	5
10	0,030	0,465	1,200	2,220	3,505	4,990	6,800	8,810	10,880	12,915	14,940	17,005	10
15	0,050	0,495	1,245	2,280	3,575	5,075	6,905	8,915	10,980	13,015	15,045	17,105	15
20	0,070	0,525	1,285	2,340	3,640	5,160	7,005	9,015	11,085	13,115	15,145	17,210	20
25	0,090	0,555	1,325	2,400	3,710	5,250	7,100	9,120	11,190	13,215	15,250	17,315	25
30	0,110	0,585	1,370	2,460	3,780	5,330	7,200	9,220	11,290	13,315	15,350	17,415	30
35	0,130	0,620	1,415	2,520	3,850	5,420	7,300	9,320	11,395	13,420	15,450	17,520	35
40	0,150	0,650	1,460	2,580	3,915	5,500	7,400	9,420	11,495	13,520	15,550	17,625	40
45	0,170	0,685	1,505	2,640	3,985	5,590	7,500	9,525	11,600	13,620	15,655	17,730	45
50	0,190	0,720	1,550	2,700	4,050	5,680	7,600	9,630	11,700	13,720	15,760	17,830	50
55	0,210	0,755	1,600	2,765	4,125	5,765	7,700	9,735	11,800	13,820	15,860	17,940	55
60	0,235	0,790	1,660	2,830	4,200	5,860	7,800	9,840	11,900	13,920	15,965	18,040	60
65	0,255	0,830	1,715	2,900	4,280	5,950	7,900	9,940	12,000	14,025	16,070	18,145	65
70	0,280	0,870	1,770	2,965	4,355	6,045	8,000	10,045	12,105	14,125	16,170	18,250	70
75	0,300	0,910	1,825	3,030	4,430	6,140	8,100	10,150	12,205	14,230	16,275	18,350	75
80	0,320	0,950	1,880	3,100	4,510	6,230	8,200	10,250	12,305	14,330	16,380	18,455	80
85	0,345	0,990	1,935	3,165	4,585	6,325	8,300	10,360	12,405	14,435	16,485	18,560	85
90	0,370	1,035	1,900	3,230	4,660	6,420	8,400	10,460	12,505	14,535	16,585	18,660	90
95	0,390	1,080	2,050	3,300	4,700	6,510	8,500	10,565	12,610	14,640	16,690	18,765	95
100	0,410	1,120	2,100	3,365	4,820	6,610	8,600	10,670	12,710	14,740	16,790	18,870	100

T°C	1200	1300	1400	1500	1600	1700	1800	1900	2000	2100	2200	x	T°C
0	18,870	20,900	22,980	25,050	27,100	29,150	30,970	32,720	34,400	35,920	37,360	x	0
5	18,970	21,000	23,080	25,150	27,205	29,250	31,055	32,810	34,475	36,000	37,420	x	5
10	19,070	21,105	23,185	25,250	27,310	29,345	31,140	32,895	34,550	36,075	37,475	x	10
15	19,175	21,210	23,290	25,350	27,415	29,425	31,230	32,970	34,625	36,150	37,530	x	15
20	19,275	21,315	23,395	25,455	27,520	29,520	31,320	33,060	34,700	36,225	37,585	x	20
25	19,380	21,420	23,500	25,555	27,625	29,615	31,405	33,145	34,780	36,300	37,635	x	25
30	19,480	21,520	23,600	25,660	27,730	29,710	31,495	33,225	34,855	36,370	37,685	x	30
35	19,580	21,625	23,705	25,765	27,830	29,800	31,580	33,310	34,930	36,450	37,735	x	35
40	19,680	21,730	23,810	25,865	27,935	29,890	31,670	33,395	35,005	36,525	37,780	x	40
45	19,780	21,830	23,910	25,970	28,040	29,980	31,755	33,475	35,080	36,600	37,830	x	45
50	19,880	21,935	24,015	26,070	28,150	30,070	31,845	33,560	35,160	36,675	37,880	x	50
55	19,980	22,040	24,120	26,175	28,255	30,160	31,930	33,650	35,235	36,750	37,925	x	55
60	20,080	22,150	24,220	26,275	28,355	30,250	32,020	33,730	35,310	36,820	37,970	x	60
65	20,180	22,250	24,320	26,380	28,450	30,340	32,110	33,815	35,390	36,900	38,015	x	65
70	20,285	22,355	24,425	26,480	28,550	30,430	32,200	33,895	35,465	36,965	38,060	x	70
75	20,385	22,460	24,530	26,585	28,650	30,520	32,285	33,980	35,540	37,035	38,100	x	75
80	20,490	22,560	24,630	26,685	28,750	30,610	32,370	34,060	35,620	37,100	38,135	x	80
85	20,590	22,665	24,735	26,790	28,850	30,700	32,460	34,150	35,695	37,170	38,170	x	85
90	20,690	22,770	24,840	26,890	28,950	30,790	32,545	34,230	35,770	37,235	38,200	x	90
95	20,795	22,875	24,945	26,995	29,050	30,880	32,630	34,315	35,845	37,300	38,235	x	95
100	20,900	22,980	25,050	27,100	29,150	30,970	32,720	34,400	35,920	37,360	38,270	x	100



W./W. 26%Ro Sheathed, 1.8 mm

emf in mv

Reference: 0°C

T°C	0	100	200	300	400	500	600	700	800	900	1000	1100	T°C
0	0,000	0,275	0,850	1,910	3,240	4,770	6,480	8,480	10,550	12,670	14,810	16,950	0
5	0,005	0,295	0,895	1,975	3,310	4,850	6,575	8,595	10,655	12,775	14,920	17,055	5
10	0,010	0,315	0,945	2,040	3,380	4,930	6,670	8,700	10,760	12,880	15,030	17,160	10
15	0,020	0,335	0,990	2,105	3,455	5,015	6,765	8,800	10,865	12,990	15,135	17,270	15
20	0,030	0,360	1,040	2,170	3,525	5,100	6,855	8,900	10,970	13,100	15,240	17,375	20
25	0,040	0,380	1,090	2,230	3,600	5,180	6,950	9,005	11,075	13,210	15,350	17,485	25
30	0,050	0,405	1,135	2,300	3,670	5,265	7,045	9,105	11,180	13,315	15,460	17,590	30
35	0,065	0,430	1,185	2,365	3,740	5,350	7,140	9,210	11,285	13,425	15,565	17,700	35
40	0,075	0,455	1,230	2,425	3,815	5,430	7,235	9,315	11,390	13,530	15,670	17,810	40
45	0,090	0,480	1,280	2,490	3,885	5,515	7,330	9,420	11,495	13,640	15,780	17,915	45
50	0,100	0,510	1,330	2,555	3,960	5,600	7,420	9,520	11,600	13,750	15,880	18,020	50
55	0,115	0,535	1,390	2,625	4,035	5,685	7,525	9,625	11,715	13,855	15,990	18,120	55
60	0,130	0,560	1,445	2,695	4,120	5,775	7,635	9,725	11,825	13,960	16,100	18,230	60
65	0,145	0,590	1,500	2,765	4,200	5,865	7,740	9,830	11,930	14,070	16,205	18,335	65
70	0,160	0,630	1,560	2,830	4,280	5,950	7,850	9,930	12,035	14,175	16,310	18,445	70
75	0,180	0,655	1,620	2,900	4,360	6,040	7,960	10,035	12,130	14,280	16,420	18,550	75
80	0,200	0,690	1,680	2,970	4,440	6,125	8,065	10,140	12,235	14,390	16,525	18,660	80
85	0,220	0,725	1,735	3,035	4,520	6,215	8,170	10,240	12,335	14,500	16,630	18,765	85
90	0,235	0,765	1,795	3,100	4,600	6,300	8,280	10,345	12,440	14,600	16,735	18,875	90
95	0,255	0,805	1,850	3,170	4,685	6,390	8,385	10,450	12,545	14,710	16,845	18,980	95
100	0,275	0,850	1,910	3,240	4,770	6,480	8,490	10,550	12,670	14,810	16,950	19,090	100

T°C	1200	1300	1400	1500	1600	1700	1800	1900	2000	2100	2200	-	T°C
0	19,090	21,260	23,410	25,560	27,730	29,895	31,920	33,750	35,450	36,900	38,020	x	0
5	19,200	21,370	23,520	25,675	27,840	30,000	32,010	33,835	35,525	36,965	38,070	x	5
10	19,310	21,480	23,625	25,780	27,945	30,100	32,105	33,920	35,600	37,020	38,120	x	10
15	19,420	21,585	23,730	25,890	28,055	30,210	32,200	34,010	35,670	37,080	38,165	x	15
20	19,530	21,690	23,840	25,995	28,165	30,315	32,300	34,100	35,750	37,140	38,215	x	20
25	19,635	21,800	23,950	26,100	28,275	30,420	32,395	34,185	35,825	37,200	38,260	x	25
30	19,745	21,905	24,055	26,210	28,380	30,525	32,490	34,270	35,900	37,260	38,310	x	30
35	19,855	22,015	24,165	26,320	28,490	30,630	32,585	34,360	35,970	37,320	38,360	x	35
40	19,965	22,120	24,270	26,425	28,600	30,740	32,680	34,450	36,040	37,380	38,410	x	40
45	20,070	22,235	24,380	26,530	28,710	30,845	32,775	34,535	36,115	37,440	38,460	x	45
50	20,180	22,340	24,490	26,640	28,820	30,950	32,870	34,620	36,200	37,500	38,510	x	50
55	20,290	22,450	24,600	26,745	28,925	31,050	32,960	34,705	36,275	37,500	38,555	x	55
60	20,400	22,555	24,705	26,850	29,030	31,145	33,045	34,790	36,345	37,605	38,600	x	60
65	20,510	22,660	24,810	26,965	29,140	31,240	33,130	34,870	36,415	37,655	38,640	x	65
70	20,620	22,770	24,920	27,075	29,250	31,335	33,220	34,955	36,485	37,710	38,685	x	70
75	20,725	22,875	25,030	27,180	29,355	31,435	33,310	35,040	36,555	37,760	38,730	x	75
80	20,830	22,980	25,135	27,290	29,460	31,530	33,395	35,120	36,625	37,810	38,775	x	80
85	20,940	23,085	25,245	27,400	29,570	31,625	33,485	35,200	36,695	37,860	38,820	x	85
90	21,050	23,195	25,350	27,510	29,685	31,720	33,570	35,285	36,765	37,915	38,865	x	90
95	21,155	23,300	25,460	27,620	29,785	31,820	33,600	35,370	36,830	37,965	38,910	x	95
100	21,260	23,410	25,560	27,730	29,895	31,920	33,750	35,450	36,900	38,020	38,950	x	100

emf in mv

T °C

2250

2000

1750

1500

1250

1000

750

500

250

W/W.26%Re

Sheathed 18 mm

35

30

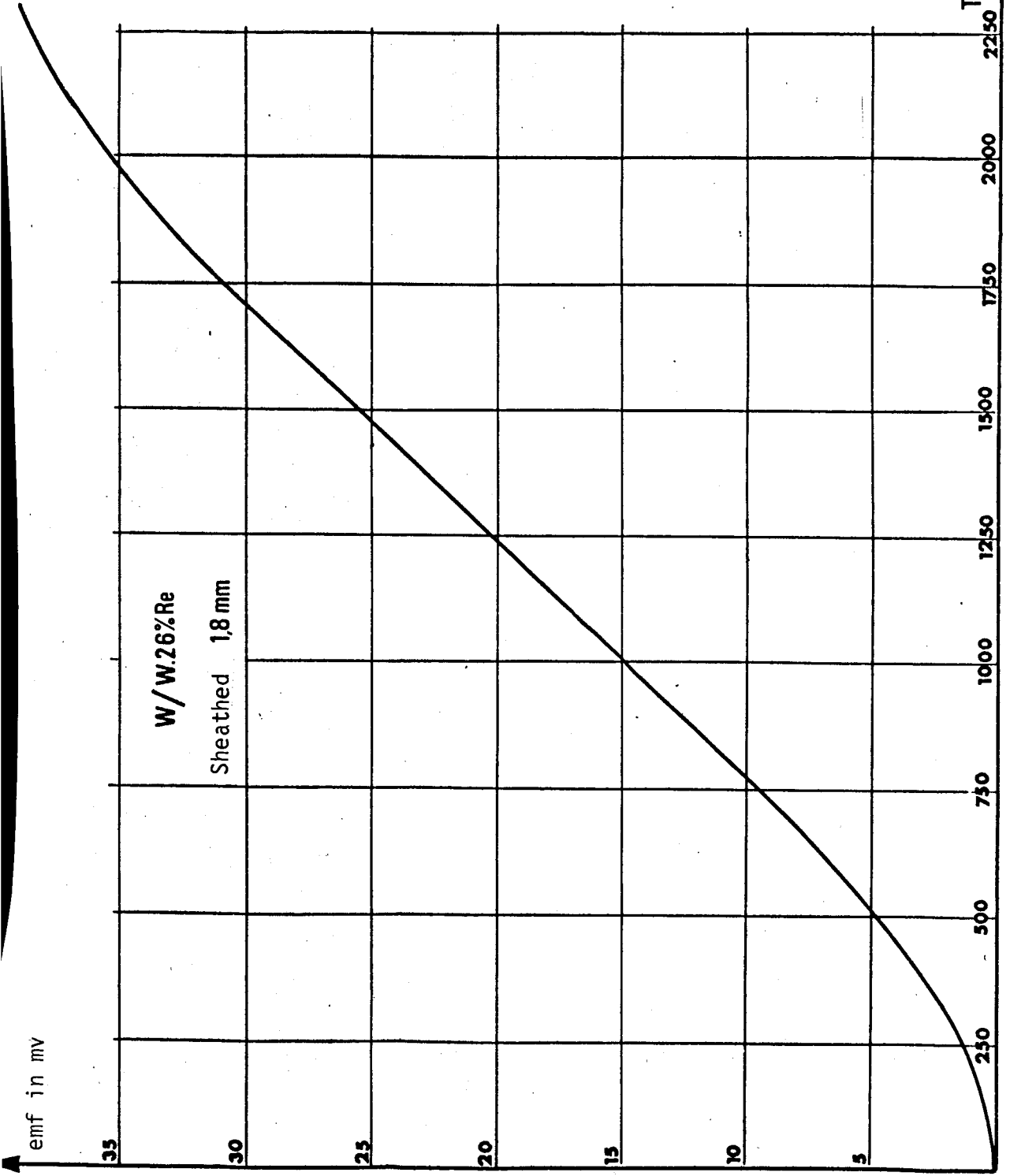
25

20

15

10

5

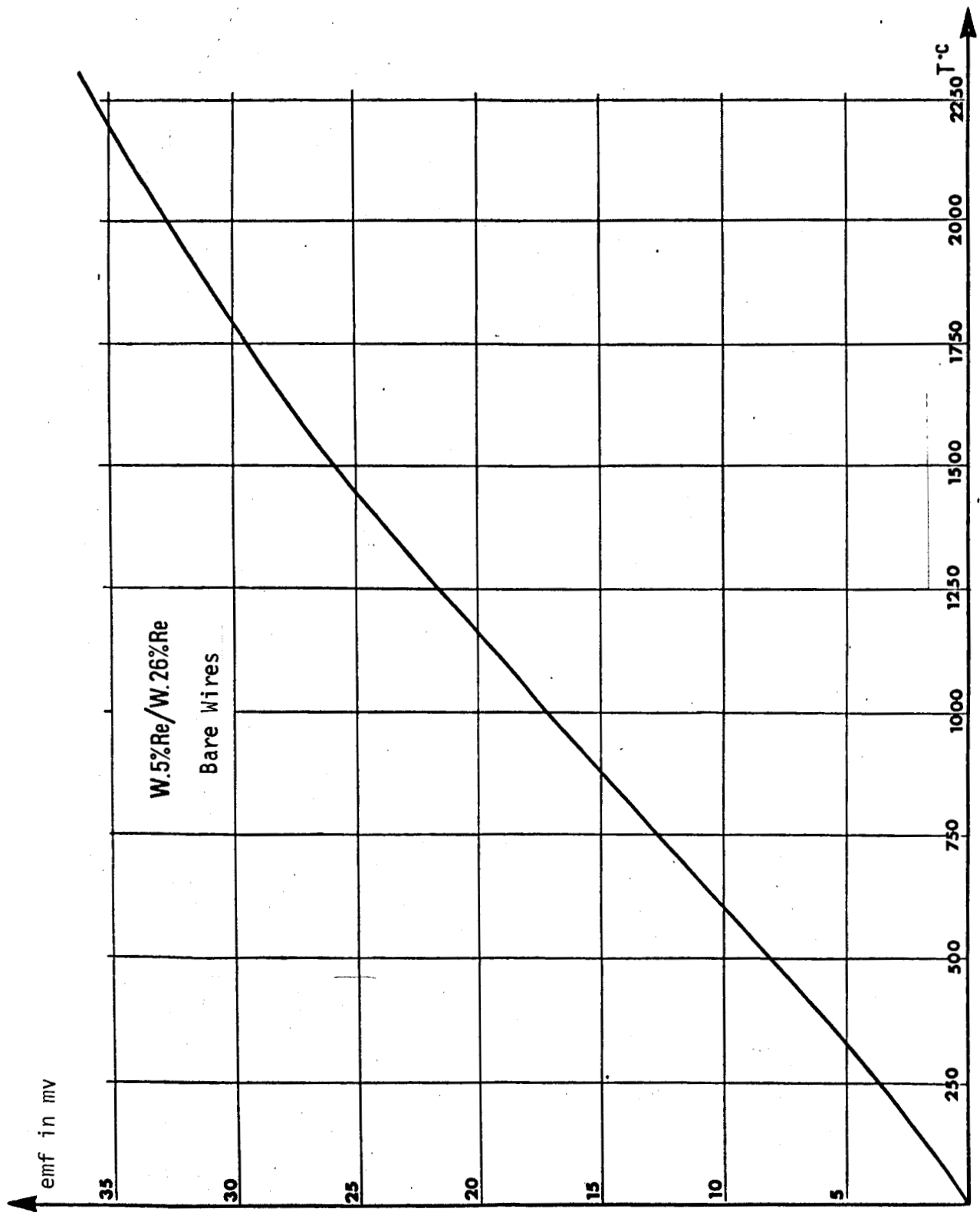


W. 5% Re / W. 26% Re Bare Wires

emf in mv
Reference: 0°C

T°C	0	100	200	300	400	500	600	700	800	900	1000	1100	T°C
0	0,000	1,370	2,870	4,550	6,360	8,180	9,980	11,780	13,580	15,380	17,180	18,985	0
5	0,065	1,460	2,950	4,640	6,455	8,270	10,070	11,870	13,670	15,270	17,270	19,070	5
10	0,130	1,515	3,035	4,730	6,545	8,360	10,160	11,960	13,760	15,560	17,360	19,160	10
15	0,200	1,585	3,115	4,820	6,635	8,450	10,250	12,050	13,850	15,650	17,450	19,250	15
20	0,270	1,660	3,200	4,910	6,725	8,540	10,340	12,140	13,940	15,740	17,540	19,340	20
25	0,340	1,730	3,280	5,000	6,895	8,630	10,430	12,230	14,030	15,830	17,630	19,430	25
30	0,405	1,805	3,360	5,090	6,905	8,720	10,520	12,320	14,120	15,920	17,720	19,515	30
35	0,475	1,880	3,440	5,180	6,995	8,810	10,610	12,410	14,210	16,010	17,810	19,600	35
40	0,540	1,950	3,525	5,270	7,085	8,900	10,700	12,500	14,300	16,100	17,900	19,690	40
45	0,610	2,025	3,610	5,365	7,175	8,990	10,790	12,590	14,390	16,190	17,990	19,780	45
50	0,680	2,100	3,690	5,455	7,265	9,080	10,880	12,680	14,480	16,280	18,080	19,870	50
55	0,750	2,175	3,780	5,545	7,355	9,170	10,970	12,770	14,570	16,370	18,170	19,960	55
60	0,820	2,250	3,865	5,640	7,455	9,260	11,060	12,860	14,660	16,460	18,260	20,045	60
65	0,885	2,330	3,950	5,730	7,535	9,350	11,150	12,950	14,750	16,550	18,350	20,135	65
70	0,955	2,405	4,035	5,820	7,625	9,440	11,240	13,040	14,840	16,640	18,440	20,225	70
75	1,025	2,485	4,120	5,910	7,715	9,530	11,330	13,130	14,930	16,730	18,530	20,310	75
80	1,090	2,560	4,200	6,000	7,810	9,620	11,420	13,220	15,020	16,820	18,620	20,400	80
85	1,160	2,635	4,290	6,090	7,900	9,710	11,510	13,310	15,110	16,910	18,710	20,490	85
90	1,230	2,715	4,375	6,180	7,990	9,800	11,600	13,400	15,200	17,000	18,800	20,580	90
95	1,300	2,790	4,460	6,275	8,085	9,890	11,690	13,490	15,290	17,090	18,890	20,665	95
100	1,370	2,870	4,550	6,360	8,180	9,980	11,780	13,580	15,380	17,180	18,985	20,755	100

T°C	1200	1300	1400	1500	1600	1700	1800	1900	2000	2100	2200	x	T°C
0	20,755	22,540	24,300	25,900	27,370	28,765	30,100	31,470	32,765	34,030	35,250	x	0
5	20,845	22,630	24,375	25,975	27,440	28,830	30,170	31,535	32,830	34,090	35,305	x	5
10	20,930	22,720	24,455	26,050	27,510	28,900	30,240	31,600	32,900	34,150	35,360	x	10
15	21,020	22,810	24,535	26,125	27,580	28,965	30,310	31,665	32,960	34,215	35,415	x	15
20	21,115	22,895	24,620	26,200	27,650	29,030	30,375	31,730	33,025	34,280	35,470	x	20
25	21,200	22,985	24,700	26,275	27,725	29,100	30,445	31,795	33,090	34,340	35,520	x	25
30	21,290	23,070	24,775	26,350	27,795	29,165	30,510	31,860	33,160	34,400	35,575	x	30
35	21,380	23,160	24,860	26,425	27,865	29,230	30,580	31,925	33,220	34,465	35,630	x	35
40	21,470	23,250	24,940	26,500	27,940	29,295	30,650	31,990	33,285	34,525	35,685	x	40
45	21,560	23,335	25,020	26,575	28,010	29,365	30,720	32,055	33,350	34,590	35,735	x	45
50	21,650	23,425	25,100	26,650	28,080	29,430	30,785	32,120	33,415	34,650	35,785	x	50
55	21,735	23,515	25,175	26,720	28,150	29,495	30,850	32,185	33,485	34,710	35,840	x	55
60	21,825	23,600	25,255	26,790	28,220	29,560	30,920	32,250	33,540	34,770	35,890	x	60
65	21,915	23,680	25,335	26,870	28,285	29,630	30,990	32,310	33,600	34,830	35,940	x	65
70	22,005	23,775	25,415	26,940	28,350	29,700	31,060	32,375	33,660	34,890	35,990	x	70
75	22,095	23,865	25,500	27,010	28,420	29,765	31,130	32,440	33,720	34,950	36,040	x	75
80	22,180	23,950	25,580	27,080	28,490	29,830	31,200	32,510	33,780	35,010	36,085	x	80
85	22,270	24,040	25,660	27,150	28,560	29,900	31,265	32,570	33,845	35,070	36,135	x	85
90	22,360	24,130	25,740	27,220	28,630	29,965	31,335	32,640	33,905	35,130	36,185	x	90
95	22,450	24,210	25,820	27,295	28,695	30,035	31,400	32,700	33,970	35,190	36,230	x	95
100	22,540	24,300	25,900	27,370	28,765	30,100	31,470	32,765	34,030	35,250	36,280	x	100



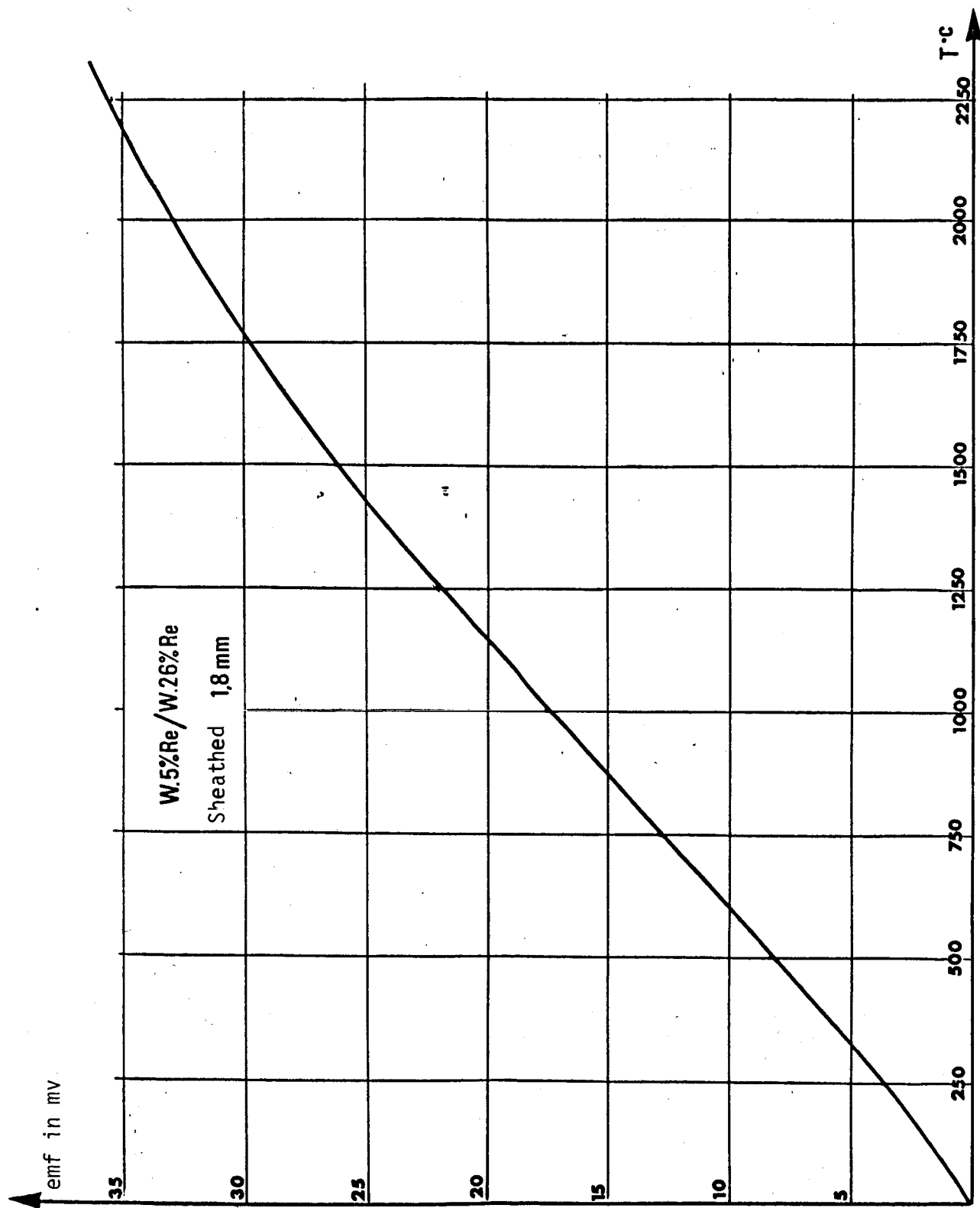
W.5% Re / W.26% Re Sheathed 1.8 mm

emf in mv

Reference: 0°C

T°C	0	100	200	300	400	500	600	700	800	900	1000	1100	T°C
0	0.000	1,250	2,810	4,600	6,440	8,275	10,115	11,950	13,790	15,630	17,465	19,305	0
5	0.060	1,320	2,895	4,690	6,530	8,365	10,205	12,040	13,880	15,720	17,555	19,395	5
10	0.115	1,390	2,985	4,785	6,625	8,460	10,300	12,135	13,975	15,815	17,650	19,490	10
15	0.170	1,460	3,075	4,875	6,715	8,550	10,390	12,225	14,065	15,905	17,740	19,580	15
20	0.225	1,535	3,165	4,970	6,810	8,645	10,485	12,320	14,160	16,000	17,835	19,675	20
25	0.280	1,610	3,255	5,060	6,900	8,735	10,575	12,410	14,250	16,090	17,925	19,765	25
30	0.340	1,685	3,345	5,150	6,990	8,825	10,665	12,500	14,340	16,180	18,015	19,855	30
35	0.400	1,760	3,435	5,245	7,085	8,920	10,760	12,595	14,435	16,275	18,110	19,950	35
40	0.460	1,840	3,525	5,335	7,175	9,010	10,850	12,685	14,525	16,365	18,200	20,040	40
45	0.520	1,920	3,615	5,430	7,270	9,100	10,945	12,780	14,620	16,460	18,295	20,135	45
50	0.585	2,000	3,705	5,520	7,360	9,195	11,035	12,870	14,710	16,550	18,385	20,225	50
55	0.650	2,080	3,795	5,610	7,450	9,285	11,125	12,960	14,800	16,640	18,475	20,315	55
60	0.715	2,160	3,885	5,705	7,545	9,380	11,220	13,055	14,895	16,735	18,570	20,410	60
65	0.780	2,240	3,970	5,795	7,635	9,470	11,310	13,145	14,985	16,825	18,660	20,510	65
70	0.845	2,320	4,060	5,890	7,730	9,565	11,405	13,240	15,080	16,920	18,755	20,605	70
75	0.910	2,400	4,150	5,980	7,820	9,655	11,495	13,330	15,170	17,010	18,845	20,690	75
80	0.975	2,480	4,240	6,070	7,910	9,745	11,585	13,420	15,260	17,100	18,935	20,780	80
85	1.040	2,560	4,330	6,165	8,005	9,840	11,680	13,515	15,355	17,195	19,030	20,875	85
90	1.110	2,640	4,420	6,255	8,095	9,930	11,770	13,605	15,445	17,285	19,120	20,965	90
95	1.180	2,725	4,510	6,350	8,190	10,025	11,865	13,700	15,540	17,380	19,215	21,055	95
100	1,250	2,810	4,600	6,440	8,275	10,115	11,950	13,790	15,630	17,465	19,305	21,145	100

T°C	1200	1300	1400	1500	1600	1700	1800	1900	2000	2100	2200	x	T°C
0	21,145	22,890	24,500	26,060	27,540	28,965	30,330	31,635	32,880	34,055	35,160	x	0
5	21,235	22,970	24,580	26,135	27,610	29,035	30,395	31,700	32,940	34,110	35,210	x	5
10	21,325	23,050	24,660	26,210	27,685	29,105	30,465	31,765	33,000	34,170	35,265	x	10
15	21,420	23,135	24,740	26,285	27,755	29,175	30,530	31,825	33,060	34,225	35,315	x	15
20	21,510	23,215	24,820	26,360	27,830	29,245	30,600	31,890	33,120	34,285	35,370	x	20
25	21,600	23,295	24,900	26,435	27,900	29,315	30,665	31,955	33,180	34,340	35,420	x	25
30	21,690	23,375	24,980	26,510	27,970	29,385	30,730	32,015	33,240	34,395	35,470	x	30
35	21,780	23,455	25,060	26,585	28,040	29,450	30,795	32,080	33,300	34,450	35,520	x	35
40	21,870	23,540	25,135	26,655	28,115	29,520	30,860	32,140	33,360	34,510	35,570	x	40
45	21,960	23,620	25,215	26,720	28,185	29,585	30,925	32,205	33,420	34,565	35,620	x	45
50	22,050	23,700	25,295	26,805	28,255	29,655	30,990	32,265	33,480	34,620	35,670	x	50
55	22,135	23,780	25,370	26,880	28,325	29,725	31,055	32,325	33,540	34,675	35,720	x	55
60	22,220	23,860	25,450	26,955	28,395	29,790	31,120	32,390	33,595	34,730	35,765	x	60
65	22,310	23,940	25,525	27,025	28,470	29,860	31,185	32,450	33,655	34,780	35,815	x	65
70	22,395	24,020	25,605	27,100	28,540	29,925	31,250	32,515	33,710	34,835	35,860	x	70
75	22,480	24,100	25,680	27,175	28,610	29,995	31,315	32,575	33,770	34,890	35,910	x	75
80	22,560	24,180	25,755	27,250	28,680	30,060	31,380	32,635	33,825	34,945	35,995	x	80
85	22,645	24,260	25,830	27,320	28,750	30,130	31,445	32,695	33,885	35,000	36,000	x	85
90	22,725	24,340	25,910	27,395	28,825	30,195	31,505	32,760	33,940	35,050	36,050	x	90
95	22,810	24,420	25,985	27,465	28,895	30,265	31,570	32,820	34,000	35,105	36,095	x	95
100	22,890	24,500	26,060	27,540	28,965	30,330	31,635	32,880	34,055	35,160	36,140	x	100



REFERENCES

General Works

1. Herzfeld: *Temperature. Its Measurement and Control in Science and Industry.*
2. Jouguet, M.: *Traité d'Electricité Théorique* (Theoretical Treatise on Electricity).
3. Nye, J.F.: *Propriétés Physiques des Cristaux* (Physical Properties of Crystals).
4. Ziman, J.M.: *Principles of Theory of Solids.*
5. Hampel: *Rare Metals Handbook.*
6. Ribaud: *Mesure des Températures* (Measurement of Temperatures).

- - - - -

7. Mott, N.F.: *Proceedings of the Royal Society*, Vol. A 156, 1936.
8. Domenicalli, C.A.: *Physical Review*, Vol. 112, No. 6, 1958.
9. Caldwell, F.R.: *National Bureau of Standards*, 1962.
10. Wilhelm, H.A., H.J. Svec, A.I. Snow, and A.H. Danne: *AECD No. 3275*, 1948.
11. Davies, D.A.: *Journal of Scientific Instruments*, Vol. 37, 1960.
12. Sims, C.T., G.B. Gaines, and R.I. Jaffee: *Review of Scientific Instruments*, Vol. 30, No. 2, 1959.
13. Lachman, J.C. and McGurty: *Symposium on Rhenium*, 1960.
14. Nadler, M.R. and C.P. Kempter: *Review of Scientific Instruments*, Vol. 32, No. 1, 1961.
15. Kuhlman, W.C.: *ASD. TDR.*, Vol. 63, p. 233, 1963.
16. Thomas, D.B.: *Journal of Research, National Bureau of Standards*, Vol. 67, C4, 1963.
17. Gross, P.H., J. Gugliotta, K.F. Krysiak, and L.B. Gross: *International Standardizing Association Transactions*, Vol. 3, No. 4, 1964.
18. Thielke, N.R. and R.L. Shepard: *High Temperature Seminar*, Oak Ridge, 1959.
19. Sanders, V.D.: *Review of Scientific Instruments*, Vol. 29, p. 917, 1958.
20. Dickinson, J.M. and L.S. Richardson: *Transactions of the American Society of Metals*, 1958.
21. Port, J.H.: "Metals for the Space Age," *Plansee Proceedings*, p. 613, 1964.
22. Kieffer, B.F., G.S. Root, and S.A. Worcester: "Metals for the Space Age," *Plansee Proceedings*, p. 571, 1964.
23. Andelin, R.L., J.D. Knight, and M. Kahn: *Transactions Met. Soc. AIME*, p. 233, 1965.
24. Kelly, N.J., W.W. Johnston, and C.D. Baumann: *Temperature. Its Measurement and Control in Science and Industry*, p. 265.
25. Browning, W.E. and C.E. Miller: *Temperature. Its Measurement and Control in Science and Industry*, p. 271.
26. Ehringer, H.: *EUR/C/4289*, No. 2, 1962.
27. *Rapport GEMP* (Report of the GEMP), 39A, 57104, 1964.
28. Shinault, L.H. and T.F. McGrath: *American Rocket Society Journal*, 1961.
29. Lachman, J.C.: *Hoskins Manufacturing Co.*, 1961.
30. Davoine F., R. Schley, and M. Villamayor: *Rapport CEA* (Report of the

- Atomic Energy Commission), R 2481, 1964.
31. *Rapport CEA* (Report of the Atomic Energy Commission), No. 2537, 1965.
 32. Wood, Van E.: *Battelle Memorial Inst.*, 1965.
 33. Hall, D.N.: *International Symposium on In-Pile Irradiation Equipment and Techn.*, Harwell, 1966, TF 6.
 34. Billington, Crawford: *Radiation Damage in Solids*, Princeton Press.
 35. Elston, J.: *Nuclear Power*, 1960.
 36. *Rapport GEMP*, 38 A 57063, p. 19, 1964.
 37. *Rapport GEMP*, 34 A 57063, p. 15, 1964.
 38. Harvey, A.: *NEI 153*, AEC, 1961.
 39. Hawkins, R.C.: *CRL 85*, AEC, 1964.
 40. *Rapport CEA*, DM/CS/CC/3952, 1966.
 41. Cusak, N. and P. Kendall: *Research Notes*, 1958.
 42. Raag, V. and H.V. Howger: *Journal of Applied Physics*, Vol. 36, No. 6, 1965.
 43. Stephenson, R.L.: *ORNL 3670*, 1965.
 44. Eremenko, V.N.: *CMIC Report 152-28*, 1961.
 45. Catteral, J.A. and S.M. Barker, "Metals for the Space Age," *Plansee Proceedings*, page 577, 1964.

(Translated for the National Aeronautics and Space Administration under Contract No. NASw-1695 by Techtran Corporation, PO Box 729, Glen Burnie, Md. 21061)